HEALTH PHYSICS

OFFICIAL JOURNAL OF THE HEALTH PHYSICS SOCIETY

Volume 6, Numbers 1/2 August, 1961

KARL Z. MORGAN
Editor-in-Chief

W. S. SNYDER J. A. AUXIER
Editors

PERGAMON PRESS NEW YORK OXFORD

HEALTH PHYSICS

The Official Journal of the Health Physics Society

EDITORS

K. Z. MORGAN (*Editor-in-Chief*), *Director, Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A.*

W. S. SNYDER, *Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A.*

J. A. AUXIER, *Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A.*

OFFICERS AND DIRECTORS OF THE HEALTH PHYSICS SOCIETY

President: JOHN S. LAUGHLIN, *Memorial Centre, 444 East 68th Street, New York* 21, *N.Y., U.S.A.*

Secretary: FRANK L. PASCHAL, JR., 5416 *Whitman Avenue, Fort Worth* 15, *Texas, U.S.A.*

Treasurer: SAUL J. HARRIS, *Atomic Accessories, Inc.*, 811 *West Merrick Road, Valley Stream, N.Y.*

Executive Secretary:* RUSSELL F. COWING, 194 *Pilgrim Road, Boston* 15, *Mass., U.S.A.*

Most Recent Past President
ELDA E. ANDERSON ✠

President-Elect
WALTER D. CLAUS

Board of Directors
G. V. BEARD
L. J. CHERUBIN
F. P. COWAN
W. T. HAM
W. H. LANGHAM
K. Z. MORGAN
R. M. SIEVERT
G. W. C. TAIT
R. C. THORBURN

* *Changes of address should be communicated to the Executive Secretary*

MEMBERS OF THE HONORARY EDITORIAL ADVISORY BOARD

Austria: J. J. ZAKOVSKY · *Belgium:* Z. M. BACQ · R. BOULENGER
Canada: F. D. SOWBY · H. E. JOHNS · G. C. BUTLER · *Denmark:* J. C. JACOBSEN · H. LEVI
France: H. P. JAMMET · L. BUGNARD · A. ALLISY · *Germany:* E. H. GRAUL · H. HOLTHUSEN
IAEA: M. SUZUKI · *India:* A. S. RAO · *Italy:* A. PERUSSIA · *Japan:* M. K. NAKAIDZUMI · S. SHIMIZU
Netherlands: W. J. OOSTERKAMP · A. H. W. ATEN, JR. · *Norway:* K. KOREN
Sweden: R. M. SIEVERT · L. CARLBOM · *Switzerland:* F. ALDER · G. JOYET · *Turkey:* P. GAMBEL
United Kingdom: W. G. MARLEY · W. BINKS · W. V. MAYNEORD · J. S. MITCHELL · E. E. POCHIN
United States: G. FAILLA · R. D. EVANS · C. L. DUNHAM · A. M. BRUES · P. S. HENSHAW · S. WARREN
J. C. BUGHER · H. A. BLAIR · L. B. SILVERMAN · L. D. MARINELLI · W. T. HAM · B. MOYER
H. M. PARKER · L. SILVERMAN · C. A. TOBIAS · C. R. WILLIAMS · S. BLOCK
Uruguay: F. LEBORGNE · *U.S.S.R.:* F. KROTKOV · *World Health Organization:* P. DOROLLE

Publishing Offices

American Continent: Pergamon Press, Inc., 122 *East* 55*th Street, New York* 22, *N.Y.*

Rest of the World: Pergamon Press Ltd., Headington Hill Hall, Oxford, England.

Published bi-monthly. Special subscription rate for members of the Health Physics Society $6 (£2. 2*s*) per annum; *for non-members, libraries, government institutions etc.* $40 (£14) per annum, *including postage.*

Single issues $9 (£2. 10*s*) *each. Back numbers* $28 (£10) per volume.

Payment must be made in advance.

PERGAMON PRESS, INC.

122 EAST 55TH STREET, NEW YORK 22, N.Y. HEADINGTON HILL HALL, OXFORD, ENGLAND

HEALTH PHYSICS

VOLUME 6 AUGUST 1961 NUMBERS 1/2

D. F. PETERSEN, V. E. MITCHELL and W. H. LANGHAM: Estimation of fast neutron doses in man by $S^{32}(n, p)P^{32}$ reaction in body hair 1

JOHN E. BALLOU and ROY C. THOMPSON: Metabolism of zinc-65 in the rat—Consideration of permissible exposure limits 6

RALPH E. THOMAS and DANIEL G. BROWN: Response of burros to neutron–γ-radiation 19

J. OKONSKI, F. W. LENGEMANN and C. L. COMAR: Incorporation of I^{131} into chicken eggs 27

R. J. CLOUTIER and MARSHALL BRUCER: Leaks in standard sealed teletherapy sources 32

C. R. RICHMOND and J. E. FURCHNER: Effect of a carbonic anhydrase inhibitor (Diamox) on Cs^{137} excretion by rats 36

D. G. BAKER, J. C. F. MACDONALD and C. G. HUNTER: A simulator for γ-ray exposure from nuclear fallout 41

H. A. KORNBERG: The use of element-pairs in radiation hazard assessment . . 46

J. DONALD LUDWICK: The analysis of plutonium-241 in urine 63

F. L. PARKER, G. D. SCHMIDT, W. B. COTTRELL and L. A. MANN: Dispersion of radiocontaminants in an estuary 66

H. F. HENRY: Problems of personnel monitoring at a criticality accident . . 86

Note:

SAUL J. HARRIS: Health and safety costs associated with the construction of a nuclear reactor 94

Health Physics Society Placement Column 96

PRINTED IN NORTHERN IRELAND AT THE UNIVERSITIES PRESS, BELFAST

HEALTH PHYSICS

VOLUME 6 OCTOBER 1961 NUMBERS 3/4

MILNER B. SCHAEFER: Some fundamental aspects of marine ecology in relation to radioactive wastes 97

DONALD W. PRITCHARD: Disposal of radioactive wastes in the ocean 103

ROBERT LOWENSTEIN: Some legal considerations in the ocean disposal of radioactive wastes . . 110

A. H. W. ATEN, JR.: Permissible concentrations of radionuclides in sea water 114

J. M. SELBY, C. A. WILLIS, B. M. BOWEN and J. H. EDGERTON: Radiation effects of a 10-MW reactor on environs of the Georgia Nuclear Laboratories 126

K. S. SOMAYAJI: Evaluation of the ventilation rate from the decay of filter-paper air samples of thoron or radon daughters 136

K. G. VOHRA, V. S. BHATNAGAR, U. C. MISHRA and S. RAGUPATHY: Measurement of caesium-137 in milk 142

J. M. SELBY, J. R. WRIGHT, B. M. BOWEN and J. H. EDGERTON: A proportional continuous river and stream water sampler at Georgia Nuclear Laboratories 149

GERALD W. KERR: Statistical treatment of environmental radio-assay data 152

R. L. ATKINSON, J. H. QUISENBERRY, A. B. MEDLEN and G. BERGER: The effect of whole body X-irradiation on the blood and other tissues of White Leghorn cockerels 163

BETTY ROSOFF, SEYMOUR RITTER, KATHLEEN SULLIVAN, HIRAM HART and HERTA SPENCER-LASZLO: Effect of chelating agents on the removal of yttrium and lanthanum from man 177

JOHN B. HURSH and ARVIN LOVAAS: The biliary excretion of radium 183

S. MARSHALL SANDERS, JR. and SARAH C. LEIDT: A new procedure for plutonium urinalysis . . 189

J. L. DONOVAN: Health physics problems of the U.S. Army Ionizing Radiation Center . . . 198

J. E. HOY: Operational experience with Kanne ionization chambers 203

Notes:

A. H. W. ATEN, JR., E. J. DE HAAN and W. M. C. DE JONG: Measurements of fast neutron flux by means of activation of phosphorus 211

NATHAN KLEIN: A broad-range chemical dosimeter 212

L. LEWIS: Pre-operational health physics training of reactor personnel 214

ROBERT B. MARTIN: Instruments for Civil Defense 216

R. D. SMITH and V. P. JOHNSON: Equipment for self-monitoring in a plutonium facility . . 218

RALPH O. WOLLAN: The role of the health physicist in a university 219

H. C. WOODS and F. M. CRANDALL: Maintenance and field performance of the PAC-G α-Survey Meter under extreme environmental conditions 223

T. G. HOBBS: Tongs used in testing for radioactive contamination 225

Letters to the Editor:

E. R. HARRIS 226

J. NEIL: Steam-detergent decontamination gun 226

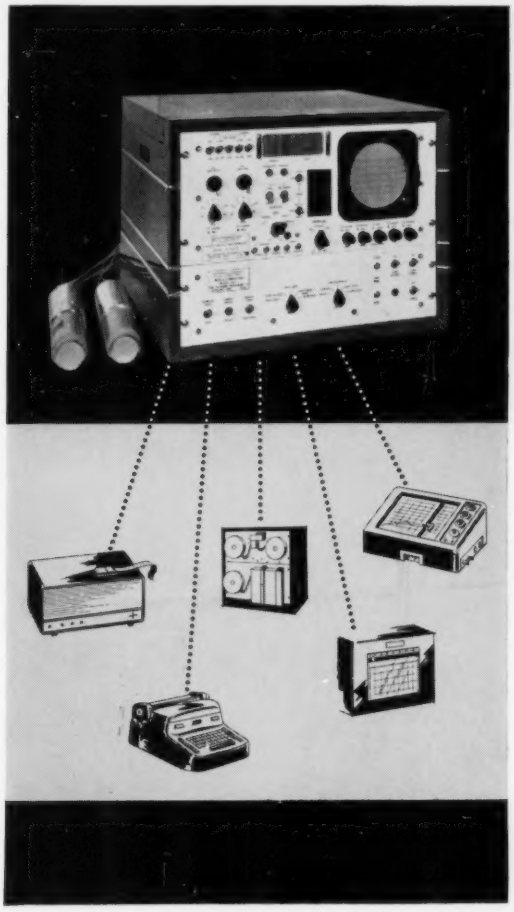
News 228

Abstracts and titles of papers presented at the Sixth Annual Meeting of the Health Physics Society . 234

A list of some of the papers to be published in future issues 253

Health Physics Society Placement Column 254

PRINTED IN NORTHERN IRELAND AT THE UNIVERSITIES PRESS, BELFAST



Now Building
MULTI-DIMENSIONAL ANALYZERS . . .

Victoreen

Victoreen, the first name in radiation measurement, is delivering the last word in multi-channel analyzers—the Tullamore-designed Model ST-200D

. . . and
400 and 800 CHANNEL
(1-8 Inputs)
"M" SERIES

- 200 or 400 Channel ferrite core memory
- Dual input—Simultaneous data accumulation
- Subgrouping and bi-directional logic
- Parallel or serial readout—Wide selection of readout devices
- Visual display and readout—Linear, log or both simultaneously
- Simultaneous linear and log plot

VICTOREEN
5806 HOUGH AVENUE • CLEVELAND 3, OHIO
EXPORT: 240 WEST 17TH ST. • NEW YORK 11, NEW YORK

YOUR ADVERTISING

will have maximum impact in

HEALTH PHYSICS

For details write or phone to

R. D. MILLER,
Advertisement Manager,

Pergamon Press Limited,
Pergamon House,
4 Fitzroy Square,
London W.1
Phone: EUSton 4455

or

Pergamon Press Inc.,
122 East 55th Street,
New York 22, N.Y.,
U.S.A.
Phone: Plaza 3-9651

THIS IS ONE OF OVER 70 PERGAMON JOURNALS! ASK FOR SPECIMENS OF OTHERS AND REDUCED RATES FOR "COMBINED" ADVERTISING.

OPPORTUNITY IN

HEALTH PHYSICS

FOR PHYSICIST, CHEMIST OR ELECTRONIC ENGINEER AT PRINCETON UNIV.

Permanent position, stimulating work in an informal research atmosphere. B.S. or M.A. Degree required with substantial work in physics and electronics. Experience helpful. Must be interested in radiation detection, instrumentation, shielding, isotope use problems.

Salary on industrial level. Unusual fringe benefits include 4 weeks paid vacation and generous retirement plan.

Write to Art Allen

ACCELERATOR

P.O. BOX 682, PRINCETON, NEW JERSEY

ADVANCED COURSE ON RADIATION PROTECTION

held at

HARWELL REACTOR SCHOOL

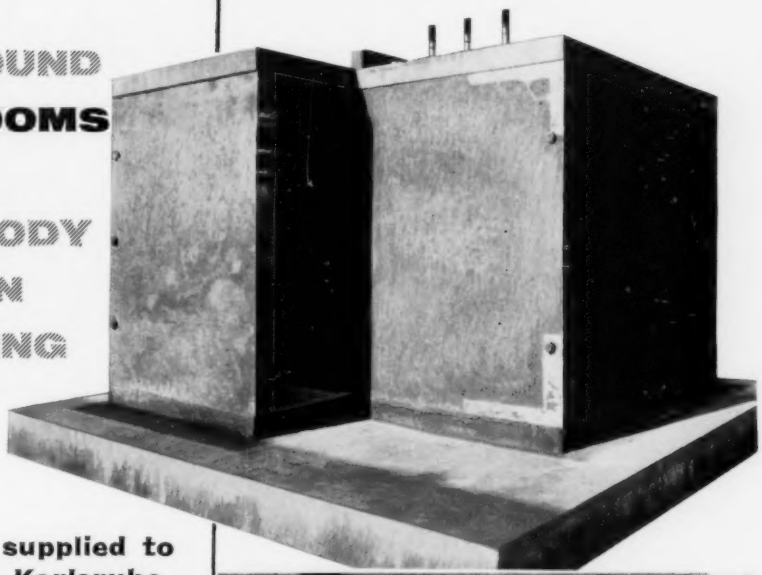
Fifth Course: 25th October 1961 to 30th January 1962

The Course is intended for persons of good academic standing who are working in or entering the field of Radiation Protection. It is especially concerned with the protection of the workers in installations having high levels of radio-activity, such as radiochemical plant, high level laboratories and reactors. The protection of the general public by control of waste products and proper transportation of radioactive material, and legal aspects also are covered. The theoretical part of the course, which includes the basic scientific principles, consists of about one hundred lectures; the majority of these are by members of the staff of the United Kingdom Atomic Energy Authority, but specialist topics are covered by members of the staffs of other bodies, including among others the Medical Research Council, the Agricultural Research Council, and the Royal Institute for Cancer Research. Practical work is held in the laboratories of the Reactor School, and in various laboratories and installations in the Atomic Energy Research Establishment. There is a comprehensive programme of visits, including a short tour of the factories of the United Kingdom Atomic Energy Authority at Windscale and Springfields.

For further information and forms of application write to:

The Manager, Reactor School,
Atomic Energy Research Establishment,
Harwell, Didcot, Berkshire.

LOW BACKGROUND STEEL ROOMS FOR HUMAN BODY RADIATION MONITORING



as already supplied to Windscale, Karlsruhe, Adelaide and Dounreay

SAUNDERS - ROE & NUCLEAR ENTERPRISES LTD.
NORTH HYDE ROAD, HAYES, MIDDLESEX. TELEPHONE: HAYES 3800

NO INTERPRETATION NEEDED!

ONE LOOK

SHOWS YOU WHETHER YOU ARE CONTAMINATED ...and WHERE!

ONLY T/A HAND & SHOE MONITORS offer this outstanding feature!

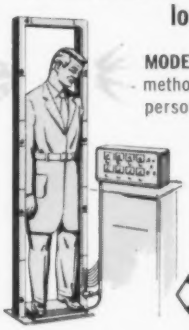
MODEL HSM-10A provides completely automatic detection and measurement of beta-gamma contamination on the back and palm of each hand and the bottom surfaces of both shoes. No skill is required in its operation.

GREATER ALARM SENSITIVITY. The Model HSM-10A employs the decade scaling principle and offers new lower warning levels adjustable from 10 to 190 counts with variable time settings from 1 to 120 seconds. An external clothing probe with rate meter read-out and speaker is included.

WRITE FOR BULLETIN NO. 167

Another T/A exclusive...

Warning Lights on T/A PORTAL MONITOR locate contamination QUICKLY!



MODEL PPM-8 provides quick, efficient method of "head to toe" monitoring of personnel entering or leaving a controlled area. It features red warning lights, one at each of the 8 detectors, which enable the user to localize quickly the area of beta/gamma contamination.

WRITE FOR BULLETIN NO. 168

180° lead shielding around each detector minimizes the background count.

LIM-18 LAUNDRY INSPECTION MONITOR

A specially designed instrument for monitoring clothing and equipment worn in beta/gamma contaminated areas. 180° lead shielding around each detector increases the sensitivity of the instrument.

T/A **TECHNICAL ASSOCIATES**

Instrumentation for Nuclear Research

140 WEST PROVIDENCIA AVENUE • BURBANK, CALIFORNIA

Heaven . . .
and
TMC
Pulse Analyzers

CN-110 256-channel Analyzer

404 400-channel Analyzer

"North *Heaven*, U.S.A." was the way our address came out in one of our ads translated for a foreign publication. Before the SEC and our competitors take us to task for claiming unfair competitive advantages, we'll admit we're a completely earthly operation. All over the earth, in fact.

Such international fame and fortune for TMC instruments comes, we think, from the universal appeal of wide usefulness in a single instrument — as exemplified by the CN-110 256-channel analyzer. It has 7 interchangeable plug-in logics, including pulse height, time of flight, pulsed neutron, multi-scaler, mass spectrometer, and coincidence pair. Circuitry is all-transistorized. Analog, binary, octal and decimal readout may be used. Data can be recorded on strip chart or X-Y recorders, printed paper tape, punched paper tape, or punched cards.

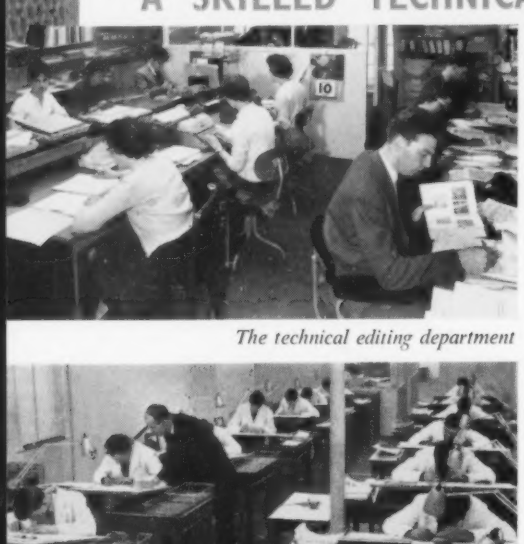
If you don't need an analyzer with this much versatility, but want instead a compact, portable, accurate, 400-channel instrument, the new TMC 404 may be just the thing for you. It has a magnetic core memory that can be used in sub groups of two or four; four separate inputs and associated amplifiers; internal pulse routing circuitry; push-button data transfer and display overlap; power requirements of *only 25 watts;* and provision for readout on a companion TMC Decimal Printer, high speed paper tape punch, strip chart or X-Y recorder, or an IBM electric typewriter.

**Note: The dog is dead . . . he had an unfortunate accident last week when struck by a suddenly swerving policy. (A temporary situation, we hope.)*

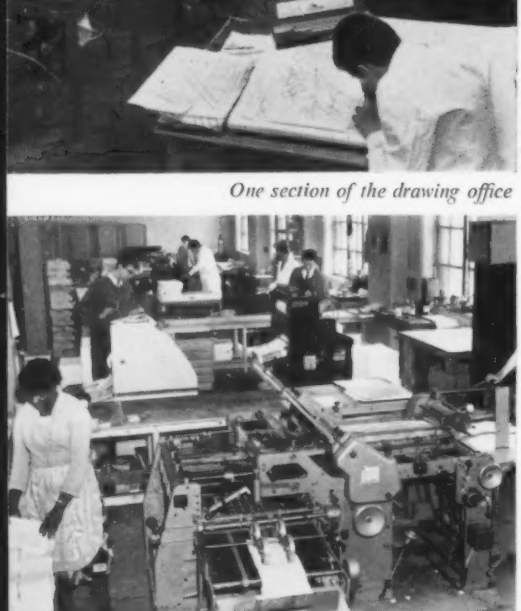
Write for literature on TMC instruments . . . to North *Haven*, Conn.

TMC **TECHNICAL MEASUREMENT CORPORATION**
441 WASHINGTON AVE., NORTH HAVEN, CONN. • CE 9-2501

A SKILLED TECHNICAL WRITING SERVICE FOR SCIENCE AND INDUSTRY



The technical editing department



One section of the drawing office

Part of the litho print room

The studio

Do you need a really fast, efficient, 'one source' technical writing, illustrating and printing service? The Pergamon Press Technical Writing Division offers a fully integrated service, capable of undertaking the complete design and production, including translation from and into foreign languages, of all types of technical printed matter, and especially user/operator handbooks, instruction and maintenance manuals, sales brochures, catalogues and company reports.

The Division is staffed by experienced, highly qualified technical writers, draughtsmen and typographic designers, and in addition can consult the several hundred internationally eminent authors and editors who publish with the Press, and obtain their advice and services in every specialized sphere of science, medicine and technology.

We offer keenly competitive prices, high quality and speedy delivery, and shall be pleased to submit quotations for all or any one of our services. We guarantee complete security when handling work of a confidential nature.

For immediate attention to your requirements please write or telephone

JOHN BURLS, B.Sc., *General Manager*

PERGAMON PRESS TECHNICAL WRITING DIVISION

566 CABLE STREET,
LONDON E.1.
Telephone: STEpney Green 4865

Editorial and sales offices also at:

4 Fitzroy Square, London W.1 Headington Hill Hall, Oxford
122 East 55th Street, New York 22, N.Y., U.S.A.
24 Rue Des Ecoles, Paris V^e^ Kaiserstrasse 75, Frankfurt am Main.

Health Physics Pergamon Press 1961. Vol. 6, pp. 1–5. Printed in Northern Ireland

ESTIMATION OF FAST NEUTRON DOSES IN MAN BY $S^{32}(n,p)P^{32}$ REACTION IN BODY HAIR*

D. F. PETERSEN, V. E. MITCHELL and W. H. LANGHAM
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico

(*Received* 3 *January* 1961)

Abstract—The high sulfur content of human hair was utilized to estimate incident fast neutron dose by measuring beta activity arising from the $S^{32}(n, p)P^{32}$ reaction. Analysis of a large number of hair samples demonstrated that regardless of variation in color, consistency, and distribution, the chemical composition was remarkably constant. The sulfur content was 47.7 ± 5.5 mg/g, and phosphorus content was 0.155 ± 0.042 mg/g of hair. Since the activation cross-sections of sulfur and phosphorus are essentially the same, virtually all P^{32} activity present in the hair sample was derived from fast neutron capture by sulfur. Experiments using the well defined fission spectrum of Godiva, an unshielded critical assembly, indicated that total neutron doses could be consistently estimated within ±10 per cent and that the orientation of samples on a plastic mannequin could be deduced by their relative specific activities. The lower limit of sensitivity of the method is approximately 5 rads of neutrons with energies in excess of 2.5 MeV using 1.0-g hair samples.

INTRODUCTION

DESPITE advances in physical dosimetry, accidents involving serious exposure to neutrons have consistently occurred in the absence of adequate physical dosimetry systems. In the earliest reported cases of HEMPELMANN and his associates,[1] neutron dose estimates were based on blood sodium activation. Subsequent incidents have similarly relied on activation measurements conducted on biological material. These procedures have been recently reviewed by HURST and RITCHIE[2]. In the most recent fatal nuclear-critical accident at the Los Alamos Scientific Laboratory, blood sodium activation provided an early indication of the magnitude of the dose,[3] and an additional biological indicator of neutron dose was also investigated which, together with additional studies on improved analytical methods, forms the basis for this report. Preliminary data obtained by this method were recently reported by HARRIS[4].

In view of the high sulfur content of hair, it was reasoned that after suitable chemical isolation, the P^{32} beta activity arising from neutron capture by sulfur in the $S^{32}(n,p)P^{32}$ reaction could be determined and the exposure to neutrons with energies in excess of 2.5 MeV computed.

MATERIALS AND METHODS

Hair, nails and cartilage occurring near the surface of the body all possess sufficient sulfur to be useful for incident fast neutron dose estimates. However, of these tissues hair is the only one readily available in sufficient quantity to permit prompt post-exposure analysis and has been used exclusively in these experiments.

Individual hair samples obtained from local barber shops and from the obstetrical and surgical services of the Los Alamos Medical Center were washed, sealed in polyethylene envelopes, and appropriately positioned on a plastic mannequin filled with tissue-equivalent solution. With the exception of two experiments conducted during the Kiwi reactor test series at the Nevada Test Site, all samples were exposed to the fission spectrum of Godiva, an unshielded Los Alamos critical assembly.

Following recovery, samples of approximately 1 g were accurately weighed and digested in 10 ml of a mixture of equal volumes of concentrated nitric and perchloric acids. Digestions were carried out in

* Work performed under the auspices of the U.S. Atomic Energy Commission.

125-ml Erlenmeyer flasks with ampoule bulb condensers. Heat was applied gradually to maintain the nitric phase of digestion for from 20 to 30 min, after which the flasks were heated to 190°C for an additional $1\frac{1}{2}$ to 2 hr. After digestion, the flasks were cooled, diluted with 15 ml of water, and brought to boiling for from 3 to 5 min. They were then diluted to approximately 50 ml with water and adjusted to pH 3.5 with 1 N ammonium hydroxide and 1 N HCl using bromphenol blue as an internal indicator. This pH adjustment was found to be critical for quantitative precipitation of phosphomolybdate. Two milligrams of carrier phosphate were added, and the P^{32} was precipitated by the addition of 25 ml of 5.5 per cent ammonium molybdate and 25 ml of 6 N nitric acid. The solution was then heated to approximately 80°C for 5 min, allowed to cool, and after standing 3 hr was filtered on a disk of Whatman no. 42 paper. The precipitate was washed with a small volume of 2 per cent nitric acid, sucked dry, and mounted on a $1\frac{1}{8}$ in. stainless steel planchet and covered with 0.5-mil Mylar. Analysis of the samples for sulfur and phosphorus was conducted on measured aliquots of the digest prior to dilution. Sulfur was measured colorimetrically with barium chloranilate by the method of LYSYJ and ZAREMBO[5]. Phosphorus was determined by the method of FISKE and SUBBAROW[6].

Throughout these studies, we have attempted to simplify the method as much as possible using readily available laboratory equipment. However, the very low activities resulting from exposure to small doses of neutrons required inordinately long counting times with conventional equipment, and a sensitive low-background plastic scintillation counter was used to check the results obtained with a thin window Geiger tube (1.6 mg/cm²) of 29.9 per cent efficiency. Dose estimates agreed quite favorably for total neutron doses in excess of 20 rads. The precipitates were counted for 30 min or to 10,000 counts. Counts were also determined at several subsequent intervals to establish the identity of the radioactive material in the sample. A typical decay curve for P^{32} was obtained on each occasion. The β-counts were corrected for counter efficiency and physical decay and expressed as dis/min per g at $T = 0$. The incident flux of sulfur-level neutrons was computed from the following equation:

$$F = \frac{A \cdot W}{\sigma \cdot s \cdot N \cdot \lambda}$$

in which F = fast neutron flux detected by sulfur (n/cm²);
A = activity/g of hair (dis/min per g at $T = 0$);
W = atomic weight of sulfur (32 g);
σ = activation cross-section of sulfur (225×10^{-27} cm²/atom);
s = sulfur content/g of hair;
N = Avogadro's number (6.02×10^{23} atoms);
λ = decay constant (3.67×10^{-5}/min).

Flux to dose conversion for neutrons of energies in excess of 2.5 MeV equals 3.83×10^{-9} rads/n per cm².[2]

RESULTS

Application of this method for estimation of the neutron dose sustained by an accident victim requires some information concerning energy distribution, since only the flux of neutrons above the sulfur threshold is derived from the computation. For the Godiva spectrum, which has been well defined,[7] neutrons above the sulfur threshold comprise approximately 30 per cent of the neutron dose, and it was therefore possible to compute total neutron doses. However, in the Kiwi reactor experiments, only fast neutron doses were calculated and compared with sulfur threshold detector data from pellets positioned at comparable distances from the device.[8]

In an attempt to simplify the procedure further, a large number of sulfur and phosphorus analyses were performed to establish whether the sulfur content of human hair varied sufficiently between individuals and with body distribution to necessitate separate elemental analysis of exposed samples. Phosphorus measurements were required to assess the potential contribution of thermal neutron activation of stable phosphorus because of the similarity of the activation cross-sections of sulfur and phosphorus.

Results shown in Table 1 indicate that the sulfur content of human hair is remarkably constant, regardless of color or body distribution. Phosphorus content was somewhat more variable, presumably due to contamination of the samples with epidermal debris. Washing materially reduced the phosphorus content in comparison with some early samples, which were not washed prior to analysis. However, no instances were observed in which phosphorus exceeded 1 per cent of the sulfur content and it was concluded that except in situations of extreme spectrum degradation, phosphorus

Table 1. Sulfur and phosphorus content of human hair

Distribution	No. samples analyzed	Sulfur mg/g mean ± s.d.	Phosphorus (mg/g) mean ± s.d.	P/S (%)
Head	64	47.1 ± 5.1	0.155 ± 0.042	0.33
Pubis	68	47.3 ± 5.6	0.251 ± 0.065	0.53
All samples (head, chest, abdomen, pubis, legs)	168	47.7 ± 5.5	—	—

activation could be disregarded as a significant source of beta activity.

A smaller series of samples was taken simultaneously from various anatomical locations of the same individual. Analytical results shown in Table 2 emphasize the constancy of chemical composition and support the idea that dose distribution can be determined by separate analysis of samples from various anatomical locations.

Data summarized in Table 3 compare total neutron doses computed from hair β-activity with estimates based on data obtained from B^{10}-clad Pu^{239}, Np^{237}, U^{238} and S^{32} threshold detectors, Hurst proportional counter, tissue-equivalent and graphite-CO_2 ionization chambers, and a calibrated fission counter.[9,10] For this series of analyses, agreement was within ±10 per cent of physical referee dosimetry.

Results of experiments in which samples were exposed during operation of the Kiwi reactors are shown in Table 4. Part of the samples were heavily contaminated with local fallout associated with operation of the reactor. However,

Table 2. Sulphur and phosphorus content of samples from various anatomical locations

Location	Sulfur (mg/g)	phosphorus (mg/g)	P/S (%)
Head	43.2	0.162	0.38
Beard	39.5	0.143	0.36
Chest	43.8	0.185	0.42
Abdomen	46.3	0.158	0.34
Pubis	45.6	0.194	0.43
Leg	46.6	0.153	0.33
Arm	48.7	0.129	0.26
Mean ± s.d.	44.8 ± 3.0	0.161 ± 0.026	0.36

Table 3. Comparison of total neutron dose estimates based on hair sulfur activation measurements with other physical dosimetry

Sample number	Hair analysis total rad dose	Referee dosimetry* total rad dose	Per cent of estimated dose
1	23	26	88
2	152	165	92
3	139	165	84
4	31	29	107
5	272	311	87
6	276	311	87
7	456	526	87
8	364	404	90
9	196	190	103
10	185	190	97
11	83	80	104
12	76	80	95
13	24	20	120
14	6	0	—
15	120	120	100
16	107	120	89
17	106	120	88
18	3	0	—
19	0	0	—
20	81	80	101

* First collision tissue dose.

Table 4. Comparison of fast neutron doses determined by hair sulfur activation and sulfur threshold detectors on mannequins exposed to neutron fluxes from the Kiwi reactors

Sample number	Hair analysis rads fast neutrons	Sulfur threshold detectors rads fast neutrons
1	77	61.3
2	82	72.0
3	8	5.4
4	6	8.3

Table 5. Dose distribution and orientation deduced from phosphorus-32 activity of irradiated hair

Sample number	Position		Total incident dose (rads)* Hair activation physical dosimetry	
	Actual	Deduced		
1	Pelvic posterior	Pelvic posterior	5	—
2	Pelvic lateral	Pelvic lateral	159	—
3	Pelvic lateral	Pelvic lateral	150	—
4	Pelvic anterior	Pelvic anterior	199	186
5	Head	Head	183	186
6	Head	Head	170	186
7	Head	Facing assembly	700	971
8	Head	Facing assembly	917	971
9	Chest	Facing assembly	905	971
10	Chest	Facing assembly	989	971
11	Pubis	Facing assembly	834	971
12	Axilla	Facing assembly	837	971

* First collision tissue dose.

in each instance, calculation of the fast neutron dose agreed favorably with the threshold detector data. These results indicate the high degree of specificity of the method and insensitivity to gross contamination with other activated products. Thus, immediate analysis of P^{32} β-activity can be performed without interference from short-lived emitters.

Table 5 shows the results of a series of experiments in which the possibility of determining dose distribution was investigated. Hair samples were placed at 90° intervals around the pelvis and on the head, or head, chest and pubis. After exposure, the analyst was required to position each sample as well as estimate the dose. The data demonstrate that accurate 180° positioning, as well as acceptable dose estimates, were achieved. However, small differences in position in essentially homogeneous radiation fields could not be discerned other than to orient the mannequin in a position facing the assembly. Similar data from hair and xyphoid cartilage measurements from the recent Los Alamos nuclear-critical accident were useful in determining the asymmetry of the fatally irradiated victim's exposure. While the head and chest received total neutron doses of approximately 2500 to 3000 rads, the pelvis due to moderation received a much smaller dose.[4]

DISCUSSION

Although these measurements are subject to greater uncertainties than the referee dosimetry, agreement was sufficient to provide a useful estimate of neutron dose with a defined spectrum. The largest discrepancies tended to occur with very low doses, but the method appears to be reliable for exposures which, in terms of total radiation, would cover a range from sublethal to supralethal doses. Moreover, information obtained after larger doses would be valuable for rapidly assessing the severity of exposure and lend support to our earlier conclusions[3] based on hair sulfur activation analysis in the Los Alamos nuclear-critical accident. The relatively constant sulfur content of hair indicates that an average value may be substituted for direct sulfur analysis, materially shortening the procedure without appreciably affecting the result.

It should be emphasized that in instances where essentially total body exposures are sustained, this method offers no advantage over blood sodium activation unless the question of orientation arises. However, sodium activation yields average values due to mixing which, under conditions of partial body shielding or gross asymmetry of the radiation field, are of little biological significance. Under these conditions, the fixed anatomical location of hair

offers the distinct advantage of providing a relative estimate of dose distribution.

The simplicity of the analytical method, the relatively small sample required, and the ready availability of hair suggest this procedure as an additional biological parameter for use in determination of first approximations of the neutron dose sustained by potential nuclear-critical accident victims.

Acknowledgments—The authors gratefully acknowledge the co-operation of Drs. J. F. SPALDING, E. R. BALLINGER, and J. A. SAYEG in exposing the samples and providing the referee neutron dosimetry.

REFERENCES

1. L. H. HEMPELMANN, H. LISCO and J. G. HOFFMAN, *Ann. Int. Med.* **36,** 279 (1952).
2. G. S. HURST and R. H. RITCHIE, Oak Ridge National Laboratory Report ORNL-2748, Part A (1959).
3. W. H. LANGHAM, *J. Occup. Med.* **3,** 169 (1961).
4. P. S. HARRIS, *Health Phys.* In press.
5. I. LYSYJ and J. E. ZAREMBO, *Microchem. J.* **3,** 173 (1959).
6. C. H. FISKE and Y. SUBBAROW, *J. Biol. Chem.* **66,** 375 (1925).
7. G. L. FRYE, J. H. GAMMEL and L. ROSEN, Los Alamos Scientific Laboratory Report LA-1670 (1954).
8. E. R. BALLINGER and D. F. PETERSEN. Unpublished observations.
9. J. A. SAYEG, E. R. BALLINGER and P. S. HARRIS, Los Alamos Scientific Laboratory Report LA-2310 (1959).
10. J. A. SAYEG, Los Alamos Scientific Laboratory Report LA-2432 (1960).

Health Physics Pergamon Press 1961. Vol. 6, pp. 6–18. Printed in Northern Ireland

METABOLISM OF ZINC-65 IN THE RAT

CONSIDERATION OF PERMISSIBLE EXPOSURE LIMITS*

JOHN E. BALLOU and ROY C. THOMPSON
Biology Laboratory, Hanford Laboratories, General Electric Company, Richland, Washington

(*Received* 4 *November* 1960; *in revised form* 3 *January* 1961)

Abstract—Data are presented on the absorption of Zn^{65} under various conditions, on its distribution and retention following a single administration, and on its buildup in tissues during chronic feeding. Highest concentrations of Zn^{65} were observed in hair, bone and prostate, the three tissues which did not attain steady-state levels of Zn^{65} during extended periods of Zn^{65} feeding. Accumulation in the total body appears to be the critical consideration, however, due to the greater absorption of the decay energy in the larger organism and the lower permissible dose rate recommended for total body irradiation. Special consideration may need be given to the very young, in view of the ready transfer of Zn^{65} across the placenta and the enhanced intestinal absorption during the first few weeks of life.

FROM a number of standpoints, Zn^{65} is a particularly interesting radioisotope. Although not a product of uranium or plutonium fission, it is a widely distributed biospheric contaminant, produced most commonly from the reaction of neutrons on stable Zn^{64}. Zinc-65 is a prominent component of the internal contamination of the Marshallese people who were exposed to the close-in fallout following explosion of a thermonuclear device at Bikini in 1954.[1] Its worldwide distribution in fallout may be inferred from its demonstrated presence in a variety of foodstuffs.[2,3] Its presence in cyclotron workers is attributed to its production by reaction of deuterons with the copper of the cyclotron dees.[4] Zinc-65 in the effluent cooling water from Hanford plutonium production reactors is specifically implicated in the detection of this radioisotope in people who drink Columbia river water downstream from the reactors, or who subsist on foodstuffs grown on land irrigated with this water.[5]

In its radioactive decay Zn^{65} exhibits rather unusual behavior, decaying predominantly by orbital electron capture, with better than 98 per cent of its energy release in the form of penetrating electromagnetic radiation. The remaining energy is released in the form of positrons, Auger electrons and weak X-rays.[6] The relatively long half-life of this radioisotope, 245 days, coupled with its ready utilization in biological systems (see VALLEE[7] for a recent review of zinc metabolism) combine to make this an isotope of potential future hazard concern.

It was our particular purpose in the present study to investigate the long term retention of Zn^{65}. The 1954 recommendations on permissible dose for internal radiation by the International Commission on Radiological Protection (ICRP) were based on an assumed biological half-life for zinc, in bone, of 23 days.[8] Subsequent studies by GILBERT and TAYLOR[9], and by PALMER[10], who detected Zn^{65} in the bones of rats fed effluent cooling water from the Hanford reactors, indicated a much more tenacious retention of zinc in bone. Results of the present investigation, together with studies of inert zinc in man,[11] and whole-body-counter data on Zn^{65} retention in man,[12,13] clearly demonstrate the long term retention of zinc. The most recent ICRP recommendations are based on biological half-lives of the order of 1000–2000 days for total body, bone and muscle.[11]

* Work performed under Contract No. AT(45-1)-1350 between the Atomic Energy Commission and the General Electric Company.

Table 1. *Effect of carrier zinc on the gastrointestinal absorption of* $Zn^{65}Cl_2$
(Retention 24 hr post feeding, per cent of dose ± s.d.)

Tissue	Carrier added (mg)			
	0	0.5	5	50
Pancreas	0.13 ± 0.002	0.082 ± 0.019	0.053 ± 0.001	0.10 ± 0.04
Spleen	0.073 ± 0.008	0.057 ± 0.019	0.025 ± 0.006	0.013 ± 0.005
Kidney	0.24 ± 0.03	0.18 ± 0.05	0.083 ± 0.015	0.053 ± 0.013
Muscle	0.18 ± 0.01	0.12 ± 0.01	0.062 ± 0.005	0.043 ± 0.009
Femur	0.072 ± 0.012	0.053 ± 0.014	0.028 ± 0.005	0.023 ± 0.006
Liver	1.2 ± 0.2	0.93 ± 0.26	0.47 ± 0.19	0.44 ± 0.10

METHODS

The rats employed in these studies were of the Sprague–Dawley strain and were maintained on Purina Laboratory Chow with free access to food and water. The radioisotope was obtained from Oak Ridge as $Zn^{65}Cl_2$ in HCl solution. The specific activity was in the range 0.4–0.7 c/g. Tissue and organ samples were analyzed for Zn^{65} content in a well-type gamma scintillation counter with a three inch sodium iodide crystal.

EXPERIMENTS AND RESULTS

Single dose gastrointestinal absorption studies

Effect of specific activity of Zn^{65}. Twelve female rats were fed by gavage, 7 μc $Zn^{65}Cl_2$ and amounts of stable zinc chloride 0, 1, 10 and 100 times the estimated daily zinc intake of 500 μg. Animals were sacrificed and assayed for Zn^{65} content 24 hr after feeding. Averaged results from the three animals in each group are summarized in Table 1. Zinc-65 absorption was only moderately influenced by substantial increases in zinc intake. This is not to say that the turnover of zinc might not be influenced by continued feeding of these high levels of zinc ingestion. Judging from these results, the very small amounts of zinc administered in other experiments reported here would not be expected to have influenced the absorption and distribution of the radioisotope. Toxic effects would not be expected at any of the zinc levels employed in these studies, since only slight toxicity has been demonstrated in rats fed diets containing as much as 0.5 per cent zinc carbonate.[14]

Effect of form of administered Zn^{65}. Since Zn^{65} is normally ingested by man in combined form in food materials, the effect of such combination on the absorbability of the radioisotope is of obvious interest. In Table 2 values are shown for the absorption of Zn^{65} following gavage of: (a) a pH 2 solution of $Zn^{65}Cl_2$, (b) a homogenized suspension of bean leaves from plants grown in a medium containing Zn^{65}, (c) a homogenized suspension of liver from a rat injected intramuscularly with Zn^{65}, and (d) and (e) suspensions of homogenized bean leaf or liver spiked with $Zn^{65}Cl_2$ solution. The rats were preconditioned to daily gavage of solutions of bean leaves or liver for 2 weeks prior to isotope administration, and were sacrificed 24 hr after a single feeding of 2 ml of the solutions containing approximately 1 μc of Zn^{65}. There were no clearly significant differences in absorption of the Zn^{65} from the various solutions,

Table 2. *The gastrointestinal absorption of* Zn^{65} *associated with plant and animal materials*
(Retention 24 hr post feeding)

	Per cent of dose ± s.d.			
	No. of animals	Liver	Carcass	Total
Solution $Zn^{65}Cl_2$, pH 2	6	3.2 ± 0.3	13 ± 1.1	16 ± 1.4
Bean leaf grown in Zn^{65} medium	4	3.1 ± 0.3	12 ± 1.9	15 ± 2.2
Liver from Zn^{65} injected rat	3	4.8 ± 1.7	18 ± 4.2	22 ± 5.3
Bean leaf spiked with $Zn^{65}Cl_2$	4	2.6 ± 0.4	10 ± 2.3	12 ± 2.7
Liver spiked with $Zn^{65}Cl_2$	4	4.2 ± 1.0	15 ± 2.2	19 ± 3.2

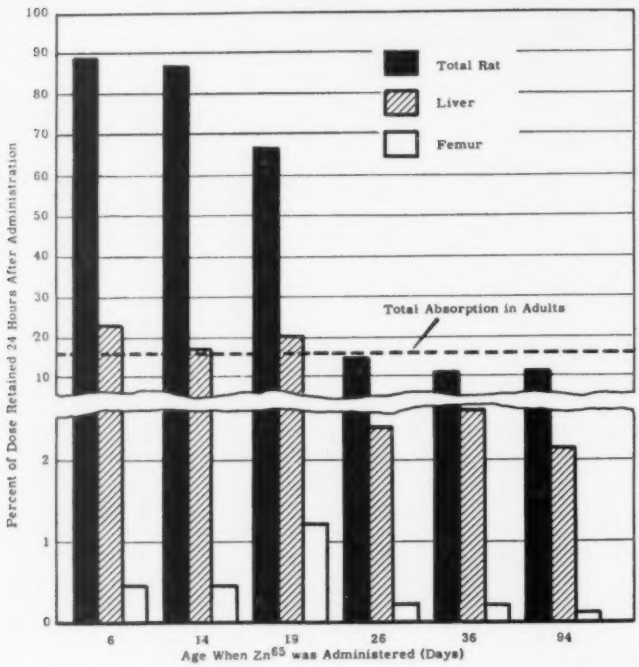


FIG. 1. Effect of age on gastrointestinal absorption of Zn^{65}.

Table 3. Effect of age on the distribution of gavage fed Zn^{65}
(Avg. ± s.d.)

Age (days)	No. of rats	Per cent of retained dose 24 hr post feeding					
		Brain	Kidney	Liver	Femur	Pelt	Carcass residue
6	4	1.1 ± 0.1	1.5 ± 0.1	26 ± 2	0.48 ± 0.01	19 ± 3	46 ± 8
14	7	1.4 ± 0.1	2.1 ± 0.2	19 ± 2	2.0 ± 0.1	24 ± 4	44 ± 2
19	7	1.5 ± 0.2	4.4 ± 0.3	30 ± 5	1.8 ± 0.2	—	55 ± 4
26	4	1.2 ± 0.3	3.1 ± 0.6	15 ± 3	1.4 ± 0.4	11 ± 3	60 ± 14
36	7	0.87 ± 0.37	4.5 ± 0.1	23 ± 4	1.6 ± 0.7	—	59 ± 18
94	4	0.54 ± 0.05	3.6 ± 0.6	18 ± 3	1.0 ± 0.2	19 ± 2	49 ± 11

although there was a consistent trend toward increased absorption in the presence of liver.

Effect of age of rat. In Fig. 1, the 24-hr retention of gavage fed $Zn^{65}Cl_2$ is shown to be markedly enhanced in rats of pre-weaning age. After weaning, at 21 days, absorption dropped abruptly to the adult level. Similar enhanced absorption has been observed in very young rats fed other radioisotopes.[15] Distribution of the retained Zn^{65}, shown in Table 3, was similar between age groups. The maximum differences in distribution were usually found between the two extremes of age studied.

Distribution and retention of Zn^{65} *following single intravenous administration*

Zinc-65 retention and distribution was determined in mature female rats for a period of 300 days following intravenous injection. The animals received 5 μc of $Zn^{65}Cl_2$ in a pH 2 solution and were sacrificed in groups of four at intervals after injection. Values for

Table 4. Retention and distribution of Zn^{65} following intravenous injection

Time after injection	1 hr	3 hr	6 hr	1 day	10 days	20 days	64 days	100 days	202 days	300 days
Avg. wt. at sacrifice (g)	224	211	230	215	211	220	228	213	246	225
Tissue	Tissue zinc-65 concentration (per cent of injected dose per g)									
Kidney	4.0	3.46	2.63	1.58	0.23	0.15	0.046	0.015	0.0027	
Heart	0.71	0.80	0.77	0.84	0.19	0.12	0.042	0.011	0.0040	
Spleen	1.95	1.88	1.72	1.38	0.21	0.13	0.034	0.013	0.0053	
Pancreas	4.60	4.84	2.44	1.59	0.28	0.18	0.031	0.012	0.0035	
Lung	0.76	0.92	0.95	0.93	0.19	0.11	0.035	0.012	0.0028	
Fat	0.097	0.093	0.095	0.092	0.040	0.014	0.0039	0.0013	0.00028	
Femur	0.56	0.61	0.64	0.56	0.64	0.50	0.42	0.38	0.28	0.24
Brain	0.095	0.11	0.10	0.16	0.16	0.14	0.040	0.014	0.0025	
Muscle	0.12	0.14	0.16	0.19	0.14	0.11	0.038	0.014	0.0023	
Liver	4.8	3.91	2.91	1.88	0.34	0.22	0.048	0.021	0.0030	
Ovaries	2.36	1.82	1.56	1.11	0.17	0.12	0.047	0.016	0.011	
Blood	0.34	0.24	0.16	0.17	0.069	0.050	0.022	0.0038		
Pelt	0.18	0.17	0.24	0.20	0.16	0.15	0.11	0.094	0.034	0.012
G.I. tract	0.81	1.17	1.0	1.20	0.14	0.10	0.023	0.008		
Total animal	0.39	0.43	0.42	0.36	0.18	0.15	0.068	0.053	0.026	0.019

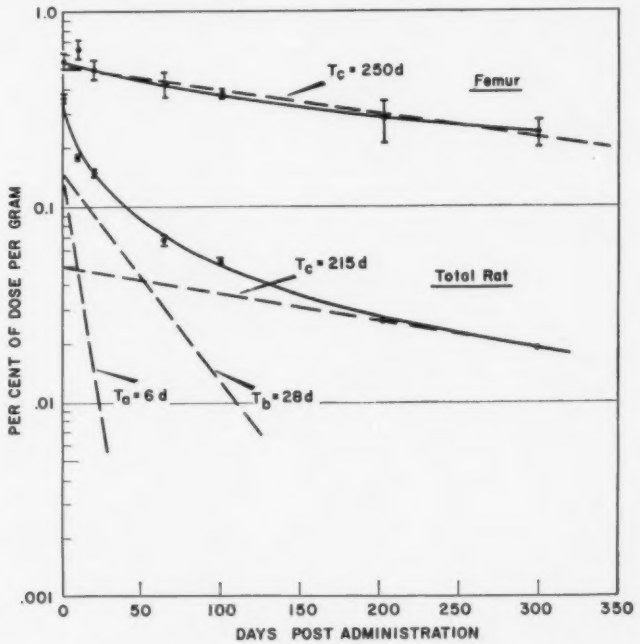


FIG. 2. Resolution of retention curves for femur and total rat (where standard deviation not indicated, pooled samples were analyzed).

retention of the radioisotope, shown in Table 4, are corrected for physical decay. Zinc-65 was measurable in only femur, pelt and carcass residue at the 300-day interval.

Retention curves for femur and total rat, Fig. 2, and for liver, Fig. 3, are shown resolved into exponential components. Retention in most tissues could not be adequately described by a single exponential

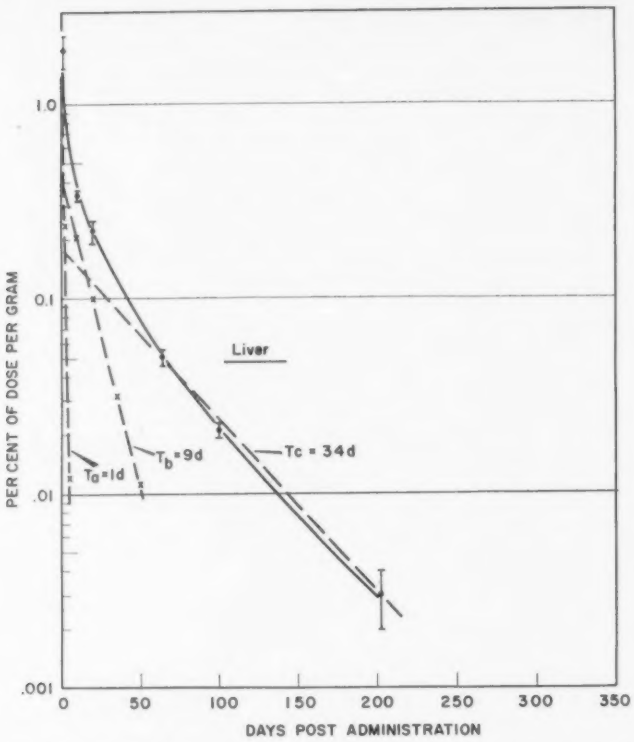


FIG. 3. Resolution of retention curve for liver (standard deviation indicated).

function nor was expression as a power function of time applicable. Exceptions were muscle and femur for which the retention data are reasonably well represented by a single exponential. The retention curve for liver illustrates the high initial uptake and rapid loss of Zn^{65} which was typical of the visceral organs studied.

The biological half-life of Zn^{65} in the various organ or tissue components, and the fraction of the total organ or tissue Zn^{65} represented by each component, are shown for several tissues in Table 5. Components with a biological half-life of 30 days or less appear to account for the bulk of the deposited radioisotope. Bone and pelt were the only tissues analyzed that retained Zn^{65} with an appreciably longer half-life. Retention in pelt was not a smooth function of time and was apparently determined largely by the hair growth cycle.

The data of Table 4 and the derived turnover rates of Table 5 invite comparison with results of a somewhat similar study in the rat reported by WAKELEY *et al.*[16]. These workers administered Zn^{65} intraperitoneally rather than intravenously and followed the retention of the radioisotope for only 42 days. Allowing for the probable effects of injection route, their data on Zn^{65} retention are in reasonable agreement with those obtained in the present study. Because of the relatively short retention period studied, their conclusions with regard to the lability of zinc in bone would appear from the present study to be erroneous. The biological half life of 15.35 days, quoted by WAKELEY *et al.*, for Zn^{65} in bone hinged very largely on the value of one experimental point at 42 days post-injection. Their further conclusion that the prostate would receive a substantially larger radiation dose from deposited Zn^{65} than would bone is not valid if the half-life of Zn^{65} in bone is as long as is indicated in the present study. Indeed, our studies of the buildup of Zn^{65} during chronic feeding (Table 7) indicate an ultimately greater accumulation of Zn^{65} in bone than in prostate.

Chronic feeding studies

Excretion of Zn^{65} *during and following chronic feeding.* Zinc-65 was administered for 200 days to female rats fed pH 2 drinking water containing 0.05 μc Zn^{65} per

Table 5. Retention of Zinc-65 in the rat following intravenous administration

Retention equation*

$$Q = Q_0\{A \exp [-(0.693t/T_a)] + B \exp [-(0.693t/T_b)] + C \exp [-(0.693t/T_c)]\}$$

Tissue	Q_0	A	T_a	B	T_b	C	T_c
Blood	0.20	0.58	3	0.42	22	—	—
Kidney	2.7	0.91	2	0.09	26	—	—
Pancreas	2.1	0.95	5	—	—	0.05	38
Spleen	2.0	0.86	2	0.14	23	—	—
Muscle	0.17	—	—	1.0	30	—	—
Liver	4.6	0.87	1	0.09	9	0.04	34
Ovaries	1.3	0.88	2	—	—	0.12	42
Femur	0.52	—	—	—	—	1.0	250
Total rat	0.35	0.43	6	0.43	29	0.14	210

* Symbols are defined as follows:

Q = concentration of Zn^{65} in tissue at time t (in days) following single administration (units of per cent of administered dose per g).

Q_0 = concentration of Zn^{65} in tissue at zero time, extrapolated from retention curve (units of per cent of administered dose per g).

A, B, C = fractions of Q_0 present at zero time in components of biological half-life T_a, T_b, T_c (in days).

ml. Results of analyses on excreta collected during and following the period of ingestion are shown in Fig. 4. Assuming that an essential balance between Zn^{65} intake and output was achieved after 200 days feeding, the observed ratio between urinary and fecal excretion would indicate that something slightly less than 2 per cent of the ingested isotope was being absorbed and excreted via the urine. Approximately thirty times as much zinc is excreted via the feces, however, as indicated by the constant ratio of urinary to fecal Zn^{65} attained after feeding of Zn^{65} was stopped. This would indicate a total absorption of approximately half of the ingested isotope.

Such an estimate is a very uncertain one, however, due to the wide fluctuations in urinary Zn^{65} output during Zn^{65} feeding, which are suggestive of possible contamination of the urine with the drinking water. In single feeding experiments, from 10 to 20 per cent of ingested zinc was absorbed and retained from 24 hr (Table 2, Fig. 1). There was no indication from the data of Table 4 of any markedly rapid turnover of zinc from the total animal during the initial 24 hr of deposition. An absorption of perhaps 25 per cent of ingested zinc, as a maximum, would therefore seem most consistent with the total observations.

Buildup of Zn^{65} *in tissues during chronic feeding.* Thirty-five female and twenty male rats, average weight 240 g and 443 g, respectively, were fed pH 2 drinking water containing 0.05 μc Zn^{65} per ml. Water consumption and therefore Zn^{65} uptake was measured periodically for each animal and remained essentially constant throughout the feeding period. Female rats were sacrificed after 45, 101, 200, 301 and 407 days on the Zn^{65} drinking regimen. Male rats were sacrificed at only the 45-, 200- and 301-day intervals.

The buildup of Zn^{65} in the female rats is shown in Table 6. These data are not corrected for radioactive decay. Steady-state concentrations were attained in most tissues long before the end of the feeding period, as would be predicted from the single exposure study. For this reason values listed for "observed buildup" are the average of concentrations observed after 200, 301 and 407 days feeding for all tissues except femur and total body. Accumulation of Zn^{65} continued in femur and hair throughout the feeding period, consistent with the slow rate of turnover of Zn^{65} in these tissues. Increasing total animal concentrations reflect the continuing buildup in bone and hair.

Also shown in Table 6 are concentrations predicted for 407 days chronic exposure, calculated from the single dose retention data. Calculations were made using the integrated form of the retention equations of Table 5, and employing a value for the fraction taken orally that arrived in the organ of interest derived as the product of the fraction retained 24 hours after gavage administration (0.16) and the fraction of total body Zn^{65} present in the organ as determined by the zero intercepts of the retention components. Use of the 24-hr retention fraction of 0.16 rather than a figure for total absorption from the gut is considered appropriate in this case because

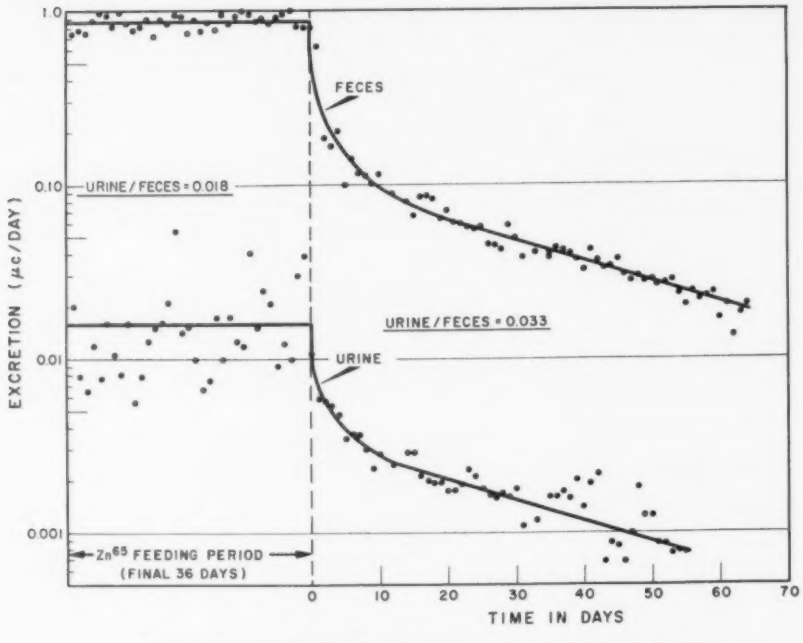


FIG. 4. Excretion of Zn^{65} during and following chronic feeding.

Table 6. *Comparison of observed and predicted buildup of* Zn^{65} *in chronically fed female rats*
(Per cent of daily dose per g)

Days chronic feeding Sacrifice weight (g)	45 247	101 239	200 229	301 254	407 264	Observed buildup	Predicted buildup
Tissues							
Brain	1.3	1.8	2.4	2.0	2.4		
Spleen	2.7	3.3	4.2	3.5	4.7	4.1	1.9
Lung	1.8	2.5	3.1	2.5	2.6		
Liver	3.9	5.2	5.8	5.8	6.0	5.9	3.0
Fat	0.23	0.31	0.49	0.39	0.45		
Kidney	3.3	4.4	4.6	4.2	4.8	4.5	2.5
Muscle	1.2	1.7	1.9	1.7	2.1	1.9	1.1
Pancreas	3.1	4.5	4.9	4.1	5.5	4.8	3.1
Femur	3.2	5.7	8.5	9.5	12	12	13
Ovaries	1.9	2.3	2.5	2.1	1.9	2.2	1.8
Heart	1.8	2.5	3.1	2.7	3.4		
Blood	0.99	0.89	0.93	0.80	1.0	0.91	0.5
Pelt	1.3	2.0	3.6	3.8	5.4		
Skin	1.1	1.5	2.1	2.3	2.7		
Hair	3.8	5.9	15	23	27		
Residual carcass	1.5	1.8	3.1	2.9	3.8		
Total rat	1.5	2.3	3.1	2.9	3.7	3.7	2.3

Table 7. Build-up of Zn^{65} in male rats during chronic feeding
(Per cent of daily dose/g tissue)

Time of feeding (days)	45	200	301
Average wt. at sacrifice (g)	432	439	386
Tissues			
Brain	0.59	1.4	1.6
Spleen	1.2	2.4	2.6
Lung	0.86	1.7	1.9
Liver	1.9	3.4	4.1
Fat	0.08	0.26	0.28
Kidney	1.4	2.2	2.7
Muscle	0.56	1.2	1.4
Pancreas	1.4	2.8	3.2
Femur	1.7	4.8	7.7
Testes	1.1	2.4	3.1
Heart	0.86	2.0	2.2
Blood	0.31	0.62	0.72
Pelt	0.78	2.3	3.5
Skin	0.54	1.2	1.8
Hair	3.2	11	14
Prostate	2.9	4.5	5.7
Residual carcass	0.49	2.1	1.9
Total rat	0.63	1.9	2.0

retention data at time intervals shorter than 1 day were not considered in the estimation of the retention equations of Table 5. The observed and predicted concentrations agree for all tissues within approximately a factor of 2.

Data on the buildup of Zn^{65} in chronically fed males is shown in Table 7. While limited as to number of sampling periods, these data show a very similar pattern of distribution to that seen with the females. The lower numerical values are, of course, a function of the unit of concentration employed and the size of the animal. Of particular interest is the continuing accumulation of Zn^{65} in the prostate. The affinity of the prostate for zinc is well known and has been widely studied.[7] In the rat, zinc concentrates in the dorsolateral lobe of the prostate,[17] with highest concentrations associated with apocrine glands in the lateral portion of this lobe.[18,19] The dorsolateral lobe of the chronically fed rats contained about 80 per cent of the total prostate Zn^{65}. In man the prostate does not exhibit a well-defined lobular structure such as is seen in the rat.[20] Areas of high zinc concentration are, however, related to that portion of the gland containing mainly secretory tissue.[21]

Retention of Zn^{65} following chronic feeding. Retention of Zn^{65} following a 200-day period of chronic oral ingestion is shown for seven tissues and organs in Fig. 5. The data are corrected for radioactive decay. The Zn^{65} was contained in the drinking water at a level of 0.05 μc/ml. The biological half-lives determined for Zn^{65} in the various organs and tissues of the rats in this experiment are very similar to those observed in the retention of a single intravenous injection (Table 5). The relative concentrations in the various tissue components are quite different in the two instances, of course, due to the buildup of Zn^{65} in components with slow turnover rates during the period of chronic exposure. The possibility of such buildup in long-lived components makes this type of experiment a sensitive check for the existence of such long-lived components that might not otherwise be observed.

Autoradiographic visualization of Zn^{65} distribution

Autoradiographs were prepared from sections taken from a rat sacrificed 3 days following a series of three daily intraperitoneal injections of 0.5 mc Zn^{65}. The autoradiographs shown in Fig. 6 are illustrative of the results obtained. A more complete account of investigations in this area of the fine structure of zinc distribution is planned for separate publication.

Placental transmission of zinc

The transfer of Zn^{65} across the placenta following the administration of single tracer doses has been previously studied by others.[7] In view of indicated changes in placental permeability during the course of gestation,[22] it was considered of interest to measure fetal Zn^{65} uptake during periods of continuous feeding of Zn^{65} to the dam. The results of limited investigations in this area are shown in Table 8. The schedules of feeding and sacrifice for the three rats and their litters are indicated in Fig. 7. The concentration of Zn^{65} attained in the litters at sacrifice was in all cases higher than the concentration in the dam. Whether this is due to a particular avidity for zinc on the part of the fetus, or is merely indicative of enhanced metabolic activity and more rapid turnover, is not clearly evident from these limited data. The fact that high levels of Zn^{65} are retained in the litter of rat *C* for a period of a week following radioisotope feeding is perhaps indicative of a high zinc demand by the very young rat. In this case, however, there was a continuing exposure of the litter via the milk from the dam. Further studies in this area are under way in our laboratory.

Consideration of permissible exposure limits

From the data obtained in these experiments

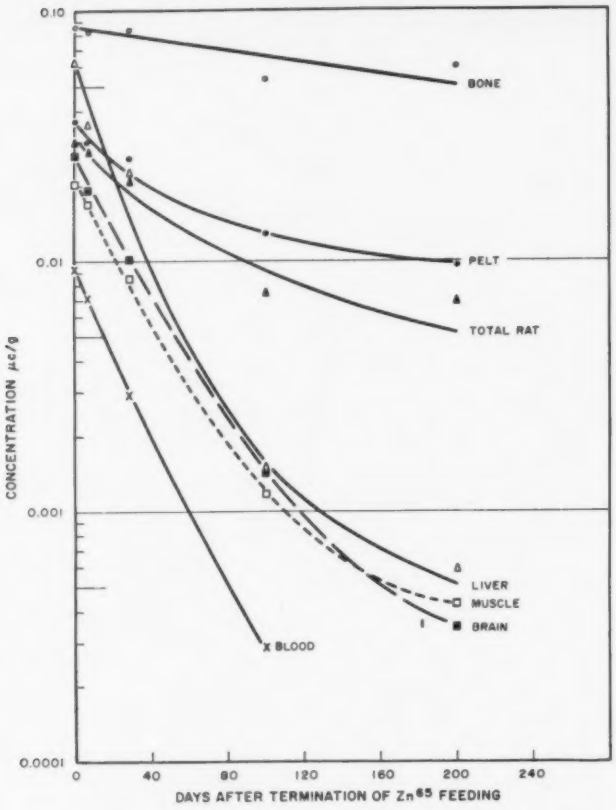


FIG. 5. Retention of Zn^{65} following chronic feeding.

Table 8. Deposition of Zn^{65} *in dam and litter of rats fed* Zn^{65} *orally during pregnancy* (expressed as μc/g tissue)

Tissue	Rat *A*		Rat *B*		Rat *C*	
	Dam	Litter	Dam	Litter	Dam	Litter
Blood	0.01	0.01	0.01	0.06	0.007	0.003
Spleen	0.07	0.03	0.05	—	—	—
Liver	0.08	0.17	0.07	0.10	0.03	0.13
Kidney	0.06	0.05	0.06	0.06	0.02	0.04
Pelt	0.01	0.03	0.01	—	0.01	0.03
Heart	0.04	0.04	0.03	0.03	—	—
Lungs	0.03	0.05	0.04	0.03	—	—
Muscle	0.01	0.03	0.01	—	—	—
Femur	0.04	0.09	0.04	0.03	0.06	0.04
Brain	0.01	0.02	0.02	0.03	—	—
Residual carcass	0.02	0.05	0.02	0.03	0.01	0.04
Total rat	0.02	0.04	0.02	0.03	0.01	0.04
Weight (g)	217	74	183	14	218	62

A. DORSOLATERAL PROSTATE (Mag.20X, NTB-3, 100μ EMULSION, 96 DAY EXPOSURE - ILLUSTRATING NON-UNIFORMITY OF DISTRIBUTION WITHIN THE LOBE.
B. KIDNEY (Mag.10X, NON-SCREEN X-RAY FILM WITH T COAT, 25μ EMULSION, 11 DAY EXPOSURE)-ILLUSTRATING PREFERENTIAL DEPOSITION IN THE CORTEX.
C. LIVER (Mag.15X, NON-SCREEN X-RAY FILM WITH T COAT, 25μ EMULSION, 12 DAY EXPOSURE)-ILLUSTRATING UNIFORM DISTRIBUTION.

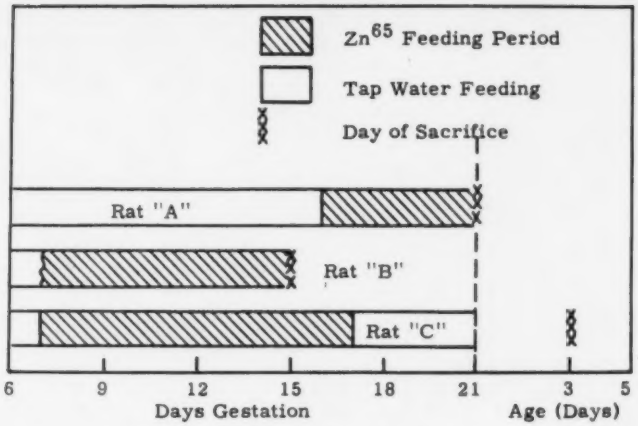
FIG. 6. Distribution of Zn^{65} as shown by autoradiographs:

A. Dorsolateral prostate ($\times 20$, NTB-3, 100 μ emulsion, 96-day exposure)—illustrating non-uniformity of distribution within the lobe.

B. Kidney ($\times 10$, non-screen X-ray film with T-coat, 25 μ emulsion, 11-day exposure)—illustrating preferential deposition in the cortex.

C. Liver ($\times 15$, non-screen X-ray film with T-coat, 25 μ emulsion, 12 day exposure)—illustrating uniform distribution.

BLANK
PAGE



Note: Dashed line indicates normal 21 day gestation period.

FIG. 7. Feeding schedule of pregnant rats.

one can calculate permissible rates of daily intake of Zn^{65} and thus permissible concentrations in water or food. Employing the procedures of the ICRP, calculations on the assumption of a number of organs as critical are outlined in Table 9. Values for the maximum permissible concentration (MPC) in water calculated from the data of the present experiment are compared with the presently recommended values of the ICRP.[11] The permissible critical organ burdens (qf_2) employed in these calculations are those calculated by the ICRP.[11] The fraction of ingested radioisotope which reaches the critical organ (fw) was calculated from the rat data on the assumption that the concentration of radioisotope in the particular organ in question would bear the same relationship to total body radioisotope concentration in man as in the rat, and that total absorption would be equivalent in man and rat. This was considered a more valid basis for extrapolation than the assumption of a constant fraction of total body Zn^{65} in the same organ of rat and man, because of the disproportionate sizes of certain organs in the two species.[23] The equation employed by the ICRP for MPC calculations assumes that the radioisotope is lost from the critical organ with an effective half-life, T. When, as in the results of the present experiments, a multiple exponential expression is required to describe radioisotope retention, the calculations become somewhat more complicated. When all half-lives are short relative to the lifetime period of exposure, this complication amounts only to the replacement of the single half-life T by the term:

$$AT_a + BT_b + CT_c + \ldots$$

where $A, B, C, \ldots$ are the fractions of the total radioisotope content present in components of effective half-life T_a, T_b, $T_c, \ldots$. The half-life values employed in the calculations of Table 9 are such "weighted average" effective half-lives, drawn directly from the rat data.

Another approach to the estimation of MPC's is shown in Table 10, where calculations are based on the results of the chronic feeding studies. In this case, one has a direct experimentally observed relationship between daily Zn^{65} intake and equilibrium levels which build up in organs and tissues. The only assumptions are those involved in adapting the rat data to man. In calculating the equilibrium concentration of Zn^{65} to be expected in man, in terms of daily intake, (C_{eq}), it was assumed that this concentration in man would be reduced from that observed in the rat by a factor of the ratio of total weight of man and rat. Thus the

Table 9. Calculation of maximum permissible concentration of zinc-65 in water from single administration data

$$\left[(\mathrm{MPC})_w = \frac{3.1 \times 10^{-4}\, qf_2}{f_w T}\right]^*$$

Organ assumed critical	qf_2 (μc)	f_w		T (days)		$(\mathrm{MPC})_w$ (μc/ml $\times 10^3$)	
		ICRP	This study	ICRP	This study	ICRP	This study
Total body	60	0.1	0.25	194	30	1	2
Liver	9.5	0.035	0.075	66	3	1	13
Kidney	2.3	0.004	0.011	93	4	2	16
Pancreas	0.7	0.003	0.003	23	6	3	12
Muscle	79	0.03	0.05	218	27	4	18
Ovaries	0.04	0.00004	0.0002	74	6	4	10
Bone	117	0.015	0.04	206	125	10	7

* This equation defines the maximum permissible concentration of a radioisotope in water when the effective half-life is short as compared with the period of exposure and when exposure is on a continuous basis.[11]

qf^2 = permissible critical organ burden as defined by ICRP (μc).
f_w = fraction of ingested radioisotope which reaches critical organ.
T = effective half-life in critical organ. (For "this study" $T = AT_a + BT_b + CT_c$, where A, B, C are fractions of total deposited radioisotope present in components of effective half-life T_a, T_b, T_c. Values are those given in Table 5, corrected to include effect of physical decay.)

Table 10. Calculation of maximum permissible concentration of zinc-65 in water from chronic administration data

$$\left[(\mathrm{MPC})_w = \frac{qf_2}{2200\, m\, C_{eq}}\right]^*$$

Organ assumed critical	qf_2 (μc)	m (g)	C_{eq} ($\times 10^4$) (fraction of daily intake/g)	$(\mathrm{MPC})_w$ (μc/ml $\times 10^3$)
Total body	60	7×10^4	1.4	3
Prostate	0.3	20	3.1	22
Liver	9.5	1.7×10^3	2.2	12
Kidney	2.3	300	1.7	20
Pancreas	0.7	70	1.8	25
Muscle	79.	3×10^4	0.7	17
Ovaries	0.04	8	0.8	28
Testes	0.2	40	1.7	13
Bone	117	7×10^3	4.5	17

* This equation defines the maximum permissible concentration of a radioisotope in water when the equilibrium level of radioisotope buildup is known and when exposure is on a continuous basis (see text).

qf_2 = permissible critical organ burden as defined by ICRP (μc).[11]
m = mass of critical organ in man (g).[11]
C_{eq} = concentration of radioisotope attained in critical organ at equilibrium (fraction of daily intake/g).
2200 = daily water intake (ml).[11]

fraction of daily intake deposited per gram of tissue which is listed in Table 10 for man is the value listed in Table 6 or Table 7, for the rat, divided by 70,000/386 for the male data and 70,000/260 for the female data. Female data were employed except in the case of prostate and testes.

The MPC values calculated by these two methods (Table 9 and 10) are in reasonable agreement with each other. They are quite significantly larger, in most instances, than the ICRP recommendations (Table 9). This is due to our measurement of a substantially more rapid turnover of Zn^{65} in most tissues of the rat than is assumed by the ICRP for man.

With the exception of those based on total body as critical organ, all of the MPC values of Tables 9 and 10, including those of the ICRP, are subject to the criticism that no account is taken of irradiation of the assumed critical organ by Zn^{65} deposited outside of this organ. Thus the permissible critical organ burden (qf_2) is calculated by the ICRP without reference to irradiation from other sources. This leads to a relatively small error for most radioisotopes, but it is a substantial one for Zn^{65} where 98 per cent of the energy release is in the form of penetrating electromagnetic radiation. WAKELEY *et al.* have calculated the radiation dosage to various tissues of the rat, taking into consideration the total body distribution of Zn^{65}.[16] MCKENNEY *et al.*, in our laboratory, have measured the difference in the radiation dose to excised and *in situ* organs of a ram previously administered Zn^{65}.[24] If one corrects for this effect, the variation in calculated MPC values for the different tissues assumed to be critical becomes predominantly a factor of the differing permissible exposure limits; 100 mrems/week in the case of total body and gonads, 300 mrems/week in the case of other soft tissues, and 560 mrems/week for bone.[11] This being the case, MPC values may be based on total body Zn^{65} retention data which may be conveniently and safely measured in man using whole body counters.[12] Limited data of this sort obtained by RICHMOND and LANGHAM[12] and by ROESCH[13], suggest an effective half-life in man of the order of 125 days, and an absorption of ingested Zn^{65} which may vary from 30–90 per cent. Such data would lead to an MPC for water from two- to five-fold lower than present ICRP recommendations.

The fact that data from so many divergent sources lead to estimates of the MPC which vary by no more than a factor of ten would seem to justify a fair degree of confidence in our present ability to evaluate the hazard of Zn^{65}. Two factors may require further consideration: (1) the possible enhancement of deposition in the fetus and new born; and (2) the possibility that, because of its biological role, Zn^{65} may concentrate very heavily in certain cells or sub-cellular fractions, to the extent that the 1–2 per cent of its energy release present in the form of short range positrons, Auger electrons and weak X-rays may play a significant role. These possibilities are being further investigated.

Acknowledgments—The authors are pleased to acknowledge the technical assistance of JOAN HESS, MARGARET LAWSON and DONNA DECHANT. Counting of Zn^{65} samples was performed by personnel of the Biological Analyses Operation under the direction of A. C. CASE. Autoradiographs were prepared by N. L. DOCKUM.

REFERENCES

1. R. A. CONARD, J. S. ROBERTSON, L. M. MEYER, W. W. SUTOW, W. WOLINS, A. LOWREY, H. C. URSCHEL, JR., J. M. BARTON, M. GOLDMAN, H. HECHTER, M. EICHER, R. K. CARVER and D. W. POTTER, *Medical Survey of Rongelap People, March 1958 Four Years After Exposure to Fallout.* U.S. Atomic Energy Commission Report BNL 534, T-135 (1959).
2. G. K. MURTHY, A. S. GOLDIN and J. E. CAMPBELL, *Science* **130,** 1255 (1959).
3. M. A. VAN DILLA, *Science* **131,** 659 (1960).
4. M. A. VAN DILLA and M. J. ENGELKE, *Science* **131,** 830 (1960).
5. R. W. PERKINS and J. M. NIELSEN, *Science* **129,** 94 (1959).
6. L. SLACK and K. WAY, *Radiations from Radioactive Atoms in Frequent Use.* U.S. Atomic Energy Commission (1959).
7. B. L. VALLEE, *Physiol. Rev.* **39,** 443 (1959).
8. Recommendations of the International Commission on Radiological Protection. *Brit. J. Radiol., Suppl.* **6,** 29 (1955).
9. I. G. F. GILBERT and D. M. TAYLOR, *Biochim. et Biophys. Acta* **21,** 545 (1956).
10. R. F. PALMER, *Hanford Biology Research-Annual Report for* 1957. HW-53500 p. 189, (1958).

11. *Report of ICRP Committee II on Permissible Dose for Internal Radiation*, 1959. *Health Phys.* **3** (1960).
12. C. R. Richmond and W. H. Langham, *Health Phys.* **2,** 97 (1959).
13. W. C. Roesch. Private communication.
14. A. Rothstein, *Toxicology of the Minor Metals.* UR-262 (1953).
15. J. E. Ballou, *Proc. Soc. Exp. Biol., N.Y.* **98,** 726 (1958).
16. J. C. N. Wakeley, B. Moffatt, A. Crook and J. R. Mallard, *Int. J. Appl. Rad. Isotopes* **7,** 225 (1960).
17. M. I. Fischer, A. O. Tikkala and C. A. Mawson, *Canad. J. Biochem. Physiol.* **33,** 181 (1955).
18. S. A. Gunn and T. C. Gould, *Proc. Soc. Exp. Biol., N.Y.* **92,** 17 (1956).
19. B. Wetterdal, *Acta Radiol. Suppl.* 156 (1958).
20. W. K. Kerr, A. G. Keresteci and H. Mayoh, *Cancer* **13,** 550 (1960).
21. C. A. Mawson and M. I. Fischer, *Canad. J. Med. Sci.* **30,** 336 (1952).
22. J. P. Feaster, S. L. Hansard, J. T. McCall and G. K. Davis, *Amer. J. Physiol.* **181,** 287 (1955).
23. R. C. Thompson, M. H. Weeks, O. L. Hollis, J. E. Ballou and W. D. Oakley, *Amer. J. Roentgenol.* **79,** 1026 (1958).
24. J. R. McKenney, R. O. McClellan and L. K. Bustad. Private communication.

Health Physics Pergamon Press 1961. Vol. 6, pp. 19–26. Printed in Northern Ireland

RESPONSE OF BURROS TO NEUTRON–γ-RADIATION*

RALPH E. THOMAS† and DANIEL G. BROWN
University of Tennessee-Atomic Energy Commission, Agricultural Research Laboratory,
Oak Ridge, Tennessee

(*Received* 6 *October* 1960)

Abstract—The response of burros to neutron–γ-radiation was studied. Seven animals received 180 rads (145 rads n, 35 rads γ) at a rate of 6 rads per min. The death of two burros at this dose level was unexpected. The clinical symptoms of these animals resembled those of central nervous system damage, heretofore seen with larger doses and much higher dose rates. The five survivors exhibited symptoms of neurological derangement but to a much lesser degree. The interval between exposure and maximum depression of leukocytes was 1 week, a shorter period than has been recorded in other burro studies. The initial rise in number of neutrophils and the depression of lymphocytes followed previously observed patterns. Epilation began as expected; however, the delayed epilation reported here is without recorded precedence in this species. There is an indication of a weight-response relationship to neutron–γ-irradiation in the burro. A comparison of the hematological response of man and burros to similar amounts of neutron–γ-radiation is presented.

INTRODUCTION

THE acute response elicited in the burro from neutron–γ-irradiation has been previously studied. In 1957, during Operation Plumbbob at the Nevada Test Site (NTS), a group of eighty-eight burros was exposed to a nuclear detonation.[1] The ratio of neutrons to γ-rays was approximately 1:1. After an accidental human radiation exposure in Oak Ridge on 16 June, 1958, burro no. 442 was exposed to a mock-up experiment in order to determine the approximate neutron dose received by the affected persons.[2,3] The neutron–γ-ray ratio during this exposure was 1:3.3.

This paper reports the response of burros during the 90-day period following exposure to neutron and γ-rays, having a ratio of 4:1, which emanated from "Godiva II", an unshielded reactor. These clinical observations were conducted in conjunction with a sodium activation study of the blood performed by the Health Physics Division of the Oak Ridge National Laboratory.

MATERIALS AND METHODS

Ten mature male burros (*Equus asinus asinus*) weighing from 310 to 476 lb were selected for the experiment. These animals had been under observation at this laboratory for a period of 5 years, therefore their health and hematological pictures were well known. The animals were airlifted to Los Alamos, New Mexico, from an elevation of 989 ft to 7200 ft, 1 week prior to the beginning of the exposures to permit partial acclimation. From 5 to 7 days after the exposures the survivors were returned by air to Oak Ridge, Tennessee, for continuing study.

While in New Mexico the animals were maintained in a corral having solid wooden sides but no overhead protection. Their diet consisted of 10 lb of good quality alfalfa hay and 1 lb of oats per day per animal. Water was provided *ad libitum*. When returned to Oak Ridge, Tennessee the survivors were stabled in an open barn and provided hay and water *ad libitum* and grain at the rate of 1 lb per animal per day. At the end of one month the animals were placed in open pasture and continued on the latter feeding regimen.

Seven burros were randomly selected for exposure

* This manuscript is published with the permission of the Director of the University of Tennessee Agricultural Experiment Station, Knoxville, Tennessee. This study was completed under Contract No. AT-40-1-GEN-242 between the University of Tennessee College of Agriculture and the Atomic Energy Commission.

† On assignment from the Veterinary Corps, United States Army. Present address: 10th General Dispensary, Frankfort, APO 757, New York, N.Y.

and three were maintained as controls. For exposure the animals were restrained in individual wooden crates. The crated burro was positioned to obtain a relatively uniform exposure to the right side. The midpoint of the torso was positioned 3 m from the center of the reactor (Fig. 1). Prior to placement for exposure the burros were tranquilized with 150–400 mg of chloropromazine which was administered intramuscularly from 30 min to 1 hr prior to the beginning of the exposure. The exposure period was 30 min.

"Godiva II"[4,5] is an unshielded assembly of uranium metal enriched to approximately 90 per cent isotopic abundance in U^{235}. The reactor is a right circular cylinder, 7 in. in diameter and about $5\frac{1}{2}$ in. in height (in foreground of Fig. 1). This device was designed to have a standardized yield of 10^{16} fissions per burst or to be operated at a lower power level. The γ-dose per burst at 1 m is approximately 30 rads with a corresponding neutron dose of 370 rads. The primary spectrum has negligible thermal and epithermal neutrons, but at distances beyond a few feet from the reactor such neutrons become significant as a result of scattering from the "Kiva"* walls and floors.

The dosimetry was performed by the personnel of the Health Physics Division of the Oak Ridge National Laboratory utilizing proportional counters[6,7] and threshold detectors[8,9] for the neutron dose. During the low-power runs the proportional counters and sulfur tablets were exposed. When the high-power (burro exposures) runs were made sulfur tablets as well as other threshold detectors were used to measure neutron dose. The activities of the sulfur tablets from the high-power runs were compared to the sulfur activation per unit dose as measured by proportional counters during the low-power exposure. This normalization yielded neutron dose in rads. Metaphosphate glasses containing silver[10,11] and chemical dosimeters[12] were utilized to measure the γ-ray dose the animals received.

Blood samples were obtained prior to exposure and 12, 24, 48 and 72 hr following the irradiation. Subsequent samples were then drawn at 1, 2, 3, 4, 6, 8 and 12 weeks. Hemoglobin determinations were obtained colorimetrically by the method of DUPRAY[13] as modified by WONG[14] and HANZEL[15]. Platelet counts were made by the phase microscopic method of BRECKER *et al.*[16]. The remainder of the hematologic values was obtained by methods described by WINTROBE[17].

* "Kiva" is the Indian name for ceremonial hut and is used for the buildings which house the critical assemblies.

RESULTS

The "exposure" dose of radiation the burros received in 30 min, as measured in air, was 145 ± 3 rads of neutrons and 35 ± 4 rads of γ-rays for a total dose of 180 rads.

General effects

Within 8 hr following the exposure there appeared swelling of the right eye lids, conjunctivitis, and a thick mucoid discharge from the right eye. Two animals also had a slight mucoid discharge from the left eye. Between 12 and 48 hr all animals lost motor control of one ear; the left ear was affected in five of the seven exposed burros. As early as 24 hr the animals appeared to be stupefied. The heads hung down and bobbed as if the animals were going to sleep and suddenly awakening. Muscular tremors over the entire body were usually apparent within 48 hr after exposure and lasted 1 or 2 days. Circling to the left was noted in two of the burros. The one which subsequently died manifested this symptom in 48 hr and the other animal on the fifth day. Two burros, one of which died, developed a paralysis of the lower lip within 48 hr. The lip hung pendulously from the chin and was immobile. Anorexia was an inconsistent observation. Some animals did not eat for 1 or 2 days while others exhibited no inappetence.

Only three animals had a temperature rise following the exposure. Two were the animals which died and the other was the most severely affected survivor. The rise was noted from 4 to 8 hr post-exposure. It did not exceed 103.7°F (normal temperature is 99–100°F). Twelve hours after irradiation the temperature had returned to normal. The non-survivors had a subnormal temperature prior to death. The abnormal lacrimation from the right eye continued to be quite profuse for about 2 weeks and then gradually diminished until the sixth week, at which time the eyes appeared normal in this respect. Auricular paralysis was still present 3 months after exposure. By the end of 2 months the surviving burros had returned to their normal weight range.

Epilation

Epilation has been observed in burros in

FIG. 1. Positioning of burros during exposure. Device in center foreground is "Godiva II".

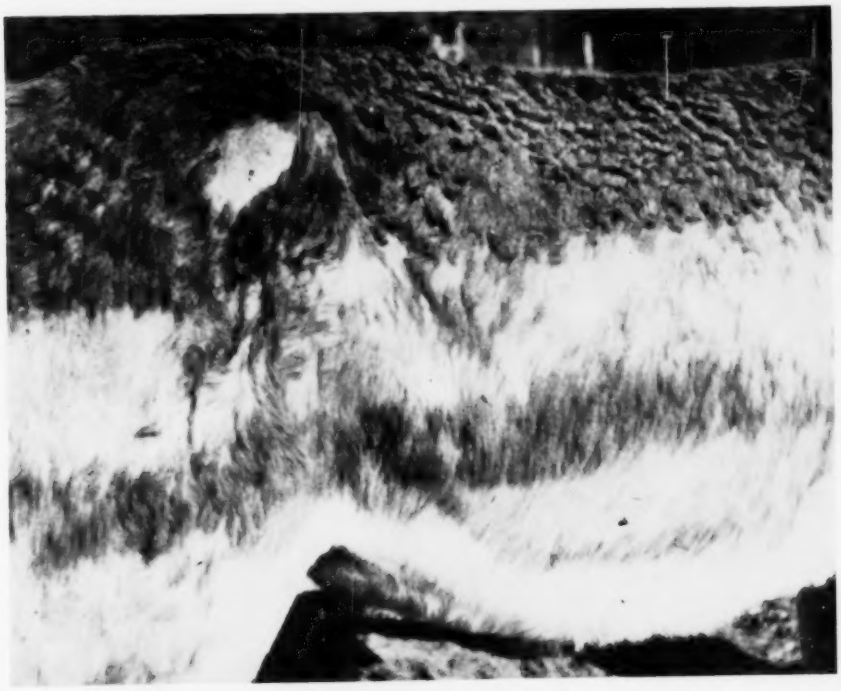


FIG. 2. Burro exhibiting stripe epilation.

FIG. 6. Burro no. 425 48 hr post-exposure.

FIG. 7. Burro no. 425 48 hr post-exposure (lateral view).

previous neutron–γ experiments to begin about the second week post-exposure. Epilation in this experiment varied greatly between time of onset and form. In view of these variations and that epilation has not been observed in burros exposed to lethal or sublethal doses of γ-radiation from Co^{60}, Ta^{182} and $Zr–Nb^{95}$, a detailed description is reported for each survivor. All epilation and loosening of hair occurred on the right side of the body, head and neck. The stripe effect created by the epilation described below is the result of shielding afforded by the boards forming the sides of the restraining crates.

Burro no. 404 showed partial epilation at 2 weeks post-exposure. It was limited to the bony prominences, i.e. over the shoulder, tuber coxae, and head of the femur. The hair pulled out easily in other areas. In 4 weeks additional epilation caused a two-stripe effect (Fig. 2). At 8 weeks the long hair ventral to the lower stripe epilated. Examination at three months revealed partial loss of tail hair. Burro no. 410 began epilating around scars and over bony prominences at 2 weeks. There was no further epilation until 9 weeks at which time stripes were faintly visible. Two stripes were prominent at 10 weeks. Three months post-exposure the last several inches of this animal's tail was completely devoid of hair. The hair of burro no. 418 was loose by the second week but no obvious epilation took place until the eighth week when a faint stripe appeared. By 9 weeks there were two definite stripes. Part of the tail hair was missing at 12 weeks. Burro no. 435 did not begin to epilate until the eighth week when one faint stripe appeared. At 9 weeks two stripes were present. By 12 weeks the tail had completely epilated. Burro no. 420 began a general epilation on the right side 2 weeks following exposure. The long outer hair shed and the undercoat remained intact. During the seventh week a stripe appeared. At 9 weeks regrowth was present. The new hair was gray in color whereas the normal coat was a chocolate brown. At 12 weeks the stripe effect was still present but there was no loss of hair from the tail.

Hematology

Total leukocytes in peripheral blood increased approximately by a factor of 2 within 12 hr and returned to pre-irradiation levels 48 hr after exposure. At 72 hr the WBC value was 66 per cent of the pre-exposure level. The lowest value (38 per cent) was reached in 7 days. Twelve weeks following exposure the leukocyte count had risen to 58 per cent of the pre-irradiation value (Fig. 3).

Prior to irradiation neutrophils accounted for 42 per cent of the white blood cells. This value rose to 90–93 per cent at 12–48 hr post-exposure, and then began a gradual decline. At 12 weeks 48 per cent of the white blood cells were neutrophils (Fig. 4). The lymphocytes dropped from a pre-irradiation level of 41 per cent to 2 per cent witin 12 hr and did not rise above 10 per cent until 1 week had elapsed. A gentle rise continued through the twelfth week at which time the lymphocytes had returned to 66 per cent of the pre-exposure value. A decrease in monocytes in the peripheral blood was noted within 24 hr. At the end of the first week the monocytes had returned to the control value.

The thrombocyte counts remained within the normal range for 1 week, after which a depression was noted. The thrombocytes reached the nadir at 3 weeks and returned to the control values between the second and third post-exposure month (Fig. 5).

Hemoglobin and hematocrit values were determined. These findings are believed to have been altered by the change in altitude without allowing adequate time for acclimation; therefore, the results are not included.[18,19]

Non-survivors

Two animals died following the exposure. Burro no. 425 died in 71 hr. Burro no. 416 died in 119 hr.

Burro no. 425 manifested lacrimation from the right eye a few hours following the exposure. At 48 hr the burro was gaunt, dyspneic and depressed. The left side of the face appeared sunken and the right side relaxed (Fig. 6). Drooping of the lower lip was evident and there was drooling of saliva. Grinding of the teeth and circling to the left were observed. When standing, the legs were spread in an effort to maintain balance (Fig. 7). These symptoms persisted for an additional 16 hr. During the next 3 hr the animal became recumbent and did

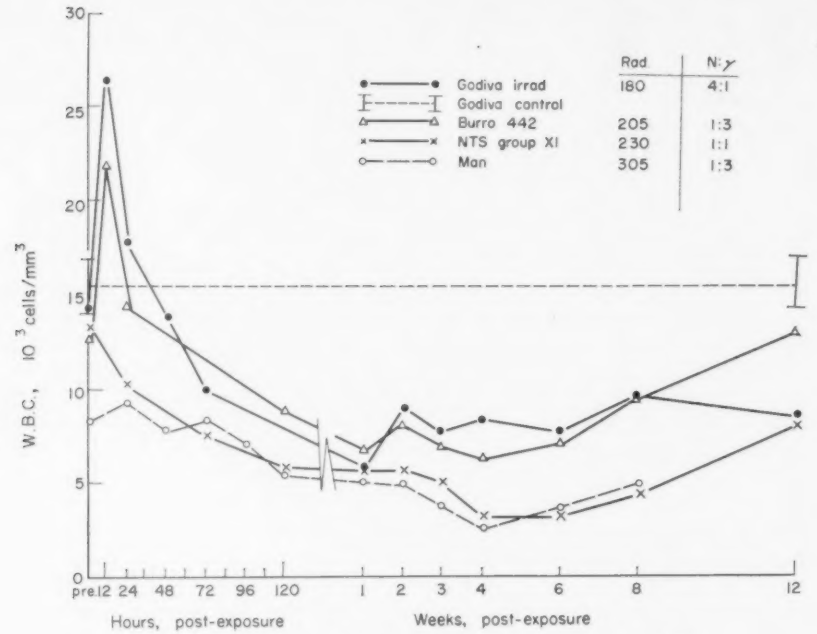


FIG. 3. Response of total leukocytes to neutron–γ-radiation.

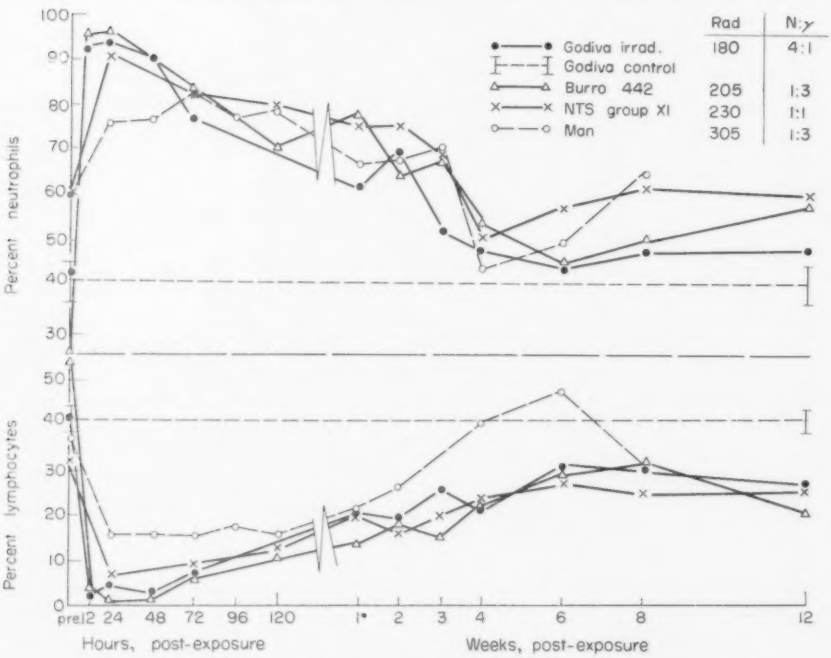


FIG. 4. Response of neutrophils and lymphocytes to neutron–γ-radiation.

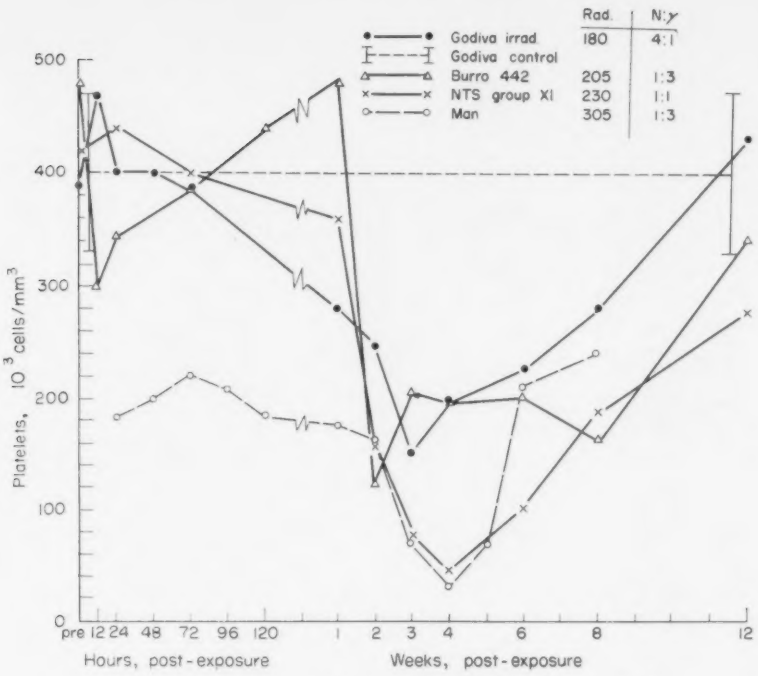


FIG. 5. Response of thrombocytes to neutron–γ-radiation.

not rise prior to its death 71 hr following exposure.

Burro no. 416 had lacrimation of the right eye within 8 hr. At 12 hr the right eye had a heavy, mucoid discharge; interference with vision was noted and the right ear drooped. No significant change was noted for the next 36 hr. Forty-eight hours after exposure the animal was eating. When the burro was observed 64 hr post-exposure, it was recumbent. Evidence in the corral indicated convulsions had been progressing for at least 4 hr. At this time muscular tremors, grinding of teeth and constant struggling were noted. These symptoms have been previously seen and attributed to central nervous system damage.[1,20] At 96 hr the animal was prostrate, cold to the touch, and unable to lift its head. The animal died 119 hr following exposure.

At necropsy the following gross lesions were noted: burro no. 425—an enlarged liver from which copious amounts of blood flowed when incised, and a soft and congested brain; burro no. 416—soft and congested brain with extensive hemorrhage at the base of the cerebellum, ecchymotic hemorrhage on serosal surface of the bladder, hematuria, and petechial hemorrhages in the subcutaneous and pericardial fat, lymph nodes and atria of the heart.

Microscopic lesions in the tissues of burro no. 425 were patchy areas of congestion, atelectasis and emphysema of the lungs, hemorrhagic areas in the pancreas, and extensive fatty change involving the liver. The brain, because of central nervous system symptoms, was examined extensively. Lesions in this organ were moderately severe congestion throughout, mild neutrophilic perivasculitis and satellitosis, and perivascular hemorrhages in the white matter of the pons.

Histopathological examination revealed a number of changes in the parenchymatous organs and tissues from burro no. 416. Hemorrhages of varying degrees were observed in the heart, lymph nodes, splenic capsule, substantia propria of the urinary bladder and in the

cerebrum (particularly involving the thalamic and cortical areas). Marked congestion was present throughout both the spleen and brain. In addition, the brain contained alterations consisting of satellitosis, apparent enlargement of the Virchow–Robin spaces, and neutrophilic perivasculitis, the latter having been demonstrated most extensively in the pons, cerebral cortex and cerebellum. Because only two animals are involved, correlations between the observed CNS symptoms and brain lesions are discussed with data of other experiments.[21]

DISCUSSION

The clinical response which was characterized by central nervous system disturbances and early mortality was similar to that previously observed in burros exposed to larger doses and much higher dose rates.[1,20] Consistent with previous studies, the prognosis was grave and the animal invariably died when the symptoms progressed as in burros no. 416 and no. 425. Mortality in burros from such a small amount of radiation has not been observed in prior studies. Previously the lowest dose of radiation causing death in burros was 250 rads of γ-radiation to the head at a dose rate of 85 rads per min to the midline of the brain.

Lesions in the central nervous system, both grossly and microscopically, were similar to those observed in the head irradiated burros and those exposed to prompt neutron–γ-radiation. Congestion and hemorrhage in the brain substance and meninges may have increased intracranial pressure which is related to the clinical behavior we have described.

The pre-exposure hematologic values compared favorably with those reported by WILDING *et al.*[22]. The response of the total leukocytes, neutrophils and lymphocytes subsequent to exposure followed the pattern observed in previous burro studies and in man.[3,23] For comparison, hematologic values of burros and man exposed to comparable dose levels of neutron–γ-radiation are plotted in Figs. 3, 4 and 5. It is noted that the maximum depression of total leukocytes occurred at 1 week, while in other studies, burro and human values were lowest at approximately 3 weeks post-exposure. The initial WBC and thrombocyte counts for man are approximately 50 per cent of the burro values which should be considered when comparing the data. It is estimated that the RBE for neutrons of the cellular elements of the peripheral blood is approximately 1, otherwise greater differences would probably have been noted as a result of the varying neutron–γ-ratios.

Epilation occurs frequently in burros following neutron exposure but is not seen in burros following γ-irradiation.[24] Epilation began during the third week in burro no. 442 (48 rads n, 158 rads γ) and in 44 per cent of the survivors of the Nevada Test Site study. In man epilation usually begins after the second week with doses as low as 300 r. The stripe pattern observed in this study is believed to have been the result of neutron shielding afforded by the wooden panels of the restraining crates. The area shielded by the panels did not epilate as extensively as did the unshielded area, thereby creating the "stripe effect" (Fig. 2). The strip of hair protected by the panel retained its natural color whereas the new hair remains unpigmented.

The epilation that occurred among the men following the accidental nuclear excursion in Oak Ridge, Tennessee had nearly ceased by the forty-fourth day. In 6 months complete regrowth had occurred. Burro no. 442 had normal hair regrowth with no apparent change in color in 3 months. At the present rate of epilation it is estimated that the "Godiva" burros will not have a normal coat for at least 9 months following exposure.

There appears to be a weight relationship to the early neutrophil change and clinical response. The maximum per cent increase of neutrophils occurred in the smallest animals (Fig. 8). The clinical response followed a similar pattern in which the smallest animals were most severely affected (Table 1). In previous studies of the acute response of three species of large animals, cattle,[25] swine and burros[26] exposed to Co^{60} γ-radiation, a weight–response relationship was not evident. KUHN *et al.*[1] observed highest mortality in the smaller burros exposed to an atomic detonation. The $LD_{50/30}$ for these animals was 435 reps and 315 reps for average weights of 370 and 260 lb, respectively.

The response of these animals to such a low dose of irradiation leads to many questions. We

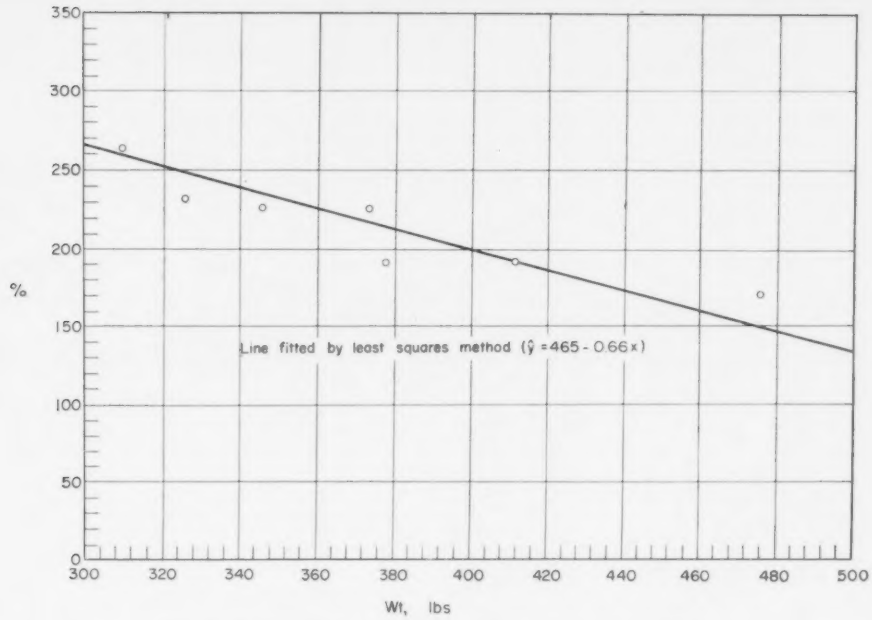


FIG. 8. Burro weight vs. maximum per cent neutrophil increase.

have seen similar responses in burros exposed to prompt neutron–γ-radiation and γ-radiation to the head at a relatively high dose rate. In each of these studies animals died from exposures of 250–280 rads which indicated a dose rate effect. However, in this study the dose rate was relatively low; thus it appears that neutrons are more effective than γ-radiation on the burro at relatively low dose rates.

Acknowledgment—The authors wish to express their appreciation to the following individuals who materially aided in this study: JOHN AUXIER and FRED SANDERS of the Oak Ridge National Laboratory; WARREN A. GRAMLEY, THOMAS G. CLARK, FANNIE H. CROSS and DANIEL P. SASMORE of this laboratory.

Table 1. *Weight–response relationship*

Animal number	Weight (lb)	Clinical response
416	310	Died
425	326	Died
410	346	Severe
418	412	Severe
435	374	Moderate
404	378	Moderate
420	476	Minimum

REFERENCES

1. U. S. G. KUHN and R. E. KYNER, *Operation PLUMBBOB, Civil Effects Test Group.* ITR 1476 (1958).
2. Union Carbide Nuclear Company, Oak Ridge, Tennessee, Report Y-1234 (1958).
3. M. BRUCER, Oak Ridge Institute of Nuclear Studies, Inc., ORINS-25 (1958).
4. *Godiva II, Its Availability and Suitability for Radiation Effects Tests,* Radiation Effects Section, Sandia Corp., Albuquerque, New Mexico (1958).
5. T. F. WINNETT and J. D. ORNDOFF, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy Geneva,* 1958, P/419. United Nations, New York (1958).
6. G. S. HURST, *Brit. J. Radiol.* **27,** 353 (1954).
7. E. B. WAGNER and G. S. HURST, *Rev. Sci. Instrum.* **29,** 153 (1958).
8. G. S. HURST, J. H. HARTER, P. N. HENSLEY, W. A. MILLS, M. SLATER, and P. W. REINHARDT, *Rev. Sci. Instrum.* **27,** 153 (1956).
9. P. W. REINHARDT and F. J. DAVIS, *Health Phys.* **1,** 169 (1958).

10. S. Davison, S. A. Goldblith and B. E. Proctor, *Nucleonics* **14,** 34 (1956).
11. J. H. Schulman, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva,* 1958, paper P/1859. United Nations, New York (1958).
12. S. C. Sigoloff, *Nucleonics* **14,** 54 (1956).
13. M. Dupray, *J. Lab. Clin. Med.* **12,** 917 (1927).
14. S. Y. Wong, *J. Biol. Chem.* **77,** 409 (1928).
15. R. F. Hanzel, *Proc. Soc. Biol. Med.* **30,** 846 (1933).
16. G. Brecker and E. P. Cronkite, *J. Appl. Physiol.* **3,** 365 (1950).
17. M. M. Wintrobe, *Clinical Hematology* (2nd Ed.) Lea and Febiger, Philadelphia (1956).
18. A. Hurtado, C. Merino and E. Delgado, *Arch. Int. Med.* **75,** 284 (1945).
19. R. E. Thomas. Unpublished data (1956).
20. *Semi-Annual Progress Report for July* 1–*December* 31, 1958, ORO-183 Agri. Res. Lab., Univ. of Tenn., Knoxville, Tenn. (1959).
21. D. G. Brown, D. P. Sasmore and L. P. Jones, The acute central nervous syndrone of burro, presented at the *International Symposium on the Response of the Nervous System to Ionizing Radiation,* Northwestern University, Chicago, Illinois, September 7–9, 1960. Proceedings to be published by Academic Press.
22. J. L. Wilding, A. W. Kimball, W. M. Whitaker, B. F. Trum and J. H. Rust, *Amer. J. Vet. Res.* **3,** 509 (1952).
23. L. H. Hamplemann, H. Lisco and J. G. Hoffman, *Ann. Int. Med.* **36,** 279 (1952).
24. B. F. Trum, J. H. Rust and J. L. Wilding, *Auburn Vet.* **3,** 131 (1952).
25. D. G. Brown and R. E. Thomas. Unpublished data (1959).
26. J. H. Rust, B. F. Trum, J. L. Wilding, C. S. Simons, C. L. Comar, *Radiology* **62,** 569 (1954).

Health Physics Pergamon Press 1961. Vol. 6, pp. 27–31. Printed in Northern Ireland

INCORPORATION OF I^{131} INTO CHICKEN EGGS*

J. OKONSKI†, F. W. LENGEMANN and C. L. COMAR
Department of Physical Biology, New York State Veterinary College,
Cornell University, Ithaca, New York

(*Received* 20 *February* 1961)

Abstract—After the Windscale accident surveys showed eggs to be the greatest source of I^{131} next to milk. Since little information is available, experiments were carried out to provide background information on the accumulation of I^{131} in eggs.

When hens received daily oral doses of I^{131} the eggs showed a plateau level of about 8 per cent of a daily dose at 6 days after the start of the experiment. Most of the I^{131} was located in the yolk, presumably in the ionic form though a significant amount may be protein bound.

With a single dose of I^{131} the maximum amount in the eggs was reached on the sixth day after dosing and indicated that maximum hazard would show a time lag.

INTRODUCTION

RADIOACTIVE iodine has been considered perhaps the most important short term hazard from nuclear debris or reactor accidents. The incorporation of I^{131} into foods other than milk has received little attention. Observations after the Windscale reactor accident indicated that eggs might be an important contributor of environmental I^{131} to the human population.[1] Therefore, the following studies were undertaken to furnish some background information on the accumulation of radioiodine in the hen's egg under typical management conditions.

PROCEDURE

Ten single-comb white Leghorn pullets of about 6 months of age were used to study the distribution of I^{131} in laying hens. The birds were kept in individual laying cages to facilitate collection of eggs and excreta. Water was supplied *ad lib.* as was a commercial laying mash (G.L.F.) that contained 2.54 mg iodine/100 g of feed. Administration of the radioisotopes to a particular bird was started only after it was established that the pullet was laying.

The first objective was to determine the levels of I^{131} that would be found in the body, excreta and eggs when the radioiodine was given daily over a long period of time. For this purpose, four of the pullets were given 1 ml of a stock I^{131} solution (10 μc per ml on day 0) every day for 19 days. The dose was placed directly in the crop of each animal by pipette and was given after measurement of body burden.

The distribution of a single oral dose of I^{131} was followed in six hens. Four of these hens received a single oral dose of 5 ml of the stock iodine solution. For comparative purposes, the other two chickens received the 5 ml dose of I^{131} injected into a wing vein.

The body content of I^{131} was determined daily by placing a hen in a small cardboard box that was located 38.1 cm away from the crystal of a scintillation probe connected to a scaler through a single channel analyzer. Each hen was then counted for 5 or 10 min. To convert the data to per cent of dose, 1 ml of the stock I^{131} solution was placed in a pint jar filled with water and counted as was the hens.

Collection of excreta were facilitated by spreading a sheet of transparent plastic on the pan under the cage of each hen. By careful folding of the sheet a complete collection of excreta could be made. The excreta were then placed in pint jars and counted in the same fashion as the hens.

* Supported in part by the U.S. Atomic Energy Commission under contract No. AT(30-1)-2147.

† Rockfeller Foundation Fellow. Present address: Polska Akademie Nauk, Poezta Grodziec Slaski, powiat Bielsko—Biata, Poland.

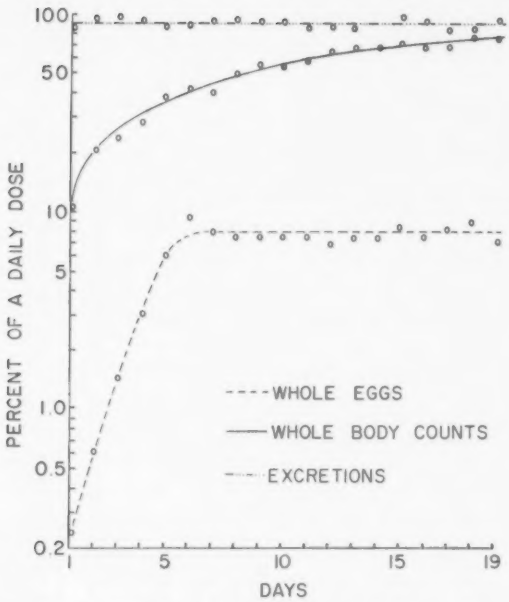


FIG. 1. The partition of daily oral doses of I^{131} between the eggs, the body, and excretions of Leghorn hens.

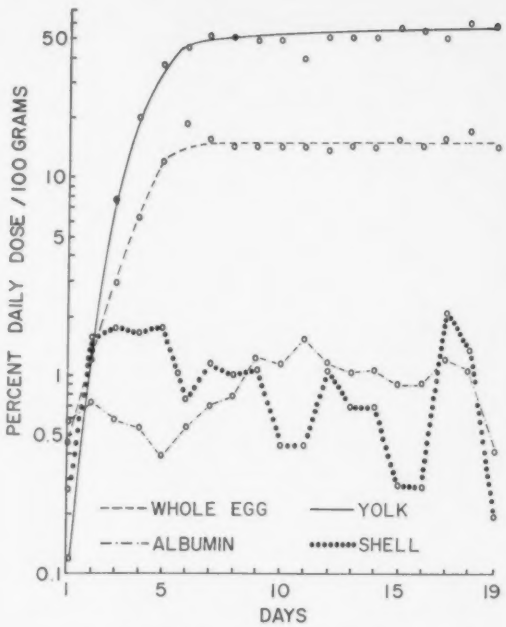


FIG. 2. The distribution of I^{131} in the eggs of hens given daily oral doses of radioiodine. The data are expressed on the basis of per cent of a daily dose per 100 g of material.

Eggs were collected every day and counted individually as the whole egg and then as yolk, albumin and shell. For counting, the eggs and the egg fractions in beakers were placed directly on the crystal of a deep-well scintillation counter. The standards were 1 ml of a 1:100 dilution of the dose or daily dose placed in beakers and brought to volumes similar to that for the albumin, yolk and shell.

At the end of the long-term experiment, the birds were killed and samples taken of muscle, liver, kidney and ovary. Fresh weights were obtained on these tissues before counting.

The data of the long-term dosing experiment are expressed as the per cent of the daily dose received by the animal; the data of the single dose experiment are expressed simply as per cent of the dose.

RESULTS

Long-term ingestion

Fig. 1 shows the accumulation of I^{131} in the bodies of the chicken, the excretion of I^{131}, and the levels found in the whole egg. To be noted is that the I^{131} content of the eggs had reached a plateau by the seventh day while the body I^{131} continued to increase for most of the experimental period. The small daily variations seen in the values for whole eggs can be explained somewhat by the fact that each hen did not lay an egg every day. It was observed that once equilibrium was reached the amount of I^{131} in the eggs of a particular hen was rather constant and did not increase if an egg had not been laid the preceding day and did not decrease if two eggs were laid on the same day. The excretion of the I^{131} doses was quite rapid approximating 85 per cent of the daily dose at the first 24-hr collection and continuing at this or a greater level throughout the experiment.

When the eggs were separated into the various parts (Fig. 2), it was noted that the yolk contained from twenty to fifty times as much I^{131} as either the albumin or the shell. Only the yolk required several days before reaching a plateau.

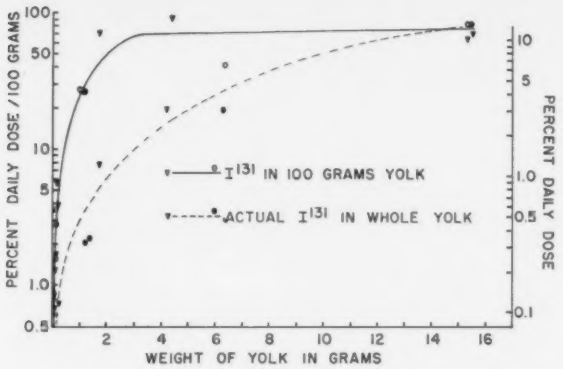


FIG. 3. The uptake of I^{131} into the yolk expressed as a function of the size of the developing ova. Triangles and circles represent ova collected from two different hens.

A preliminary fractionation of the yolk material revealed that little I^{131} was associated with the ether extractable portion. A major amount of the yolk I^{131} was extractable with water and then easily adsorbed on an ion exchange resin Dowex I-8X. This presumably is iodide I^{131}. A significant portion of the I^{131} did appear to be associated with the proteins of the yolk. Further examination of the distribution of radioiodine in the yolk is in progress.

At sacrifice muscle, liver, kidney and ovary tissue samples were found to contain, respectively, 0.04, 0.10, 0.19 and 0.52 per cent of a daily dose per 100 g of tissue. In measuring the ovary all ova larger than 0.5 cm in diameter were removed.

With two of the birds, different sized ova were collected, weighed, counted, and the data expressed as per cent of a daily dose per 100 g (Fig. 3). It was noted that the smallest ova contained the least I^{131} per unit of weight; as the ova increased in weight up to 2 g the rate of I^{131} accumulation was comparatively high. After 2 g, the I^{131} increased roughly in proportion to the gain in weight of the follicle. It should be noted, however, that most of the I^{131} was accumulated during the period when the weight of the ova increased from 2 to 16 g.

Table 1 contains a summary of the values obtained at or near equilibrium for the various samples taken. A slight difference is seen between the values for whole egg and for the fractions of the egg. This difference presumably is attributable to counting geometry.

Single-dose studies

The route of administration, oral or intravenous, made little difference in the handling of the I^{131} and so the data of all single-dose experiments were composited. A very rapid excretion of a radioiodine was noted after the administration of a single dose (Fig. 4). As with the multiple-dose study, approximately 90 per cent of the dose was excreted within the first 24 hr, after this, the amount excreted was very low and extremely variable. The body content of I^{131} reflected the losses through excretion and egg laying.

Table 1. *Summary of* I^{131} *content of various samples at or near equilibrium*

Tissue	% of daily dose	% of daily dose per 100 g
Whole body	70 ± 2.8	—
Feces (daily)	90 ± 4.5	—
Whole eggs (daily)	8 ± 0.7	15 ± 1.4
Yolk (daily)	9 ± 0.9	52 ± 5.2
Albumin (daily)	0.3 ± 0.03	1.1 ± 0.3
Shell (daily)	0.07 ± 0.04	1.0 ± 0.5

1. For whole body the values from 14–19 days were used in obtaining these values. For other tissues, the data from the following periods were used: feces, 2–19; whole egg, 7–19; yolk, 7–19; albumin, 2–19; shell, 2–19 days.

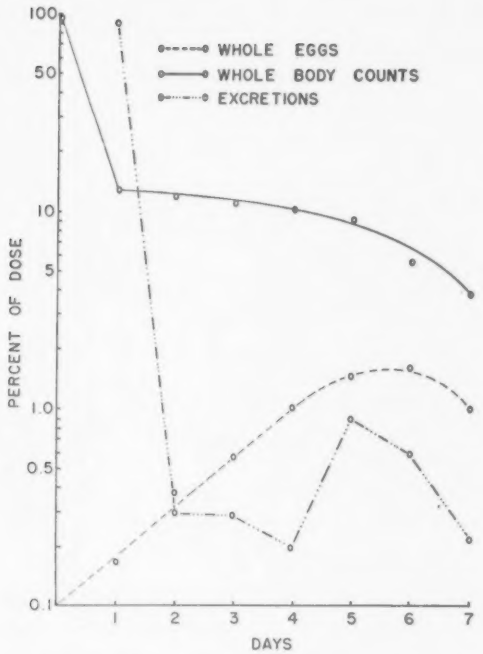


FIG. 4. The partition of I^{131} into the eggs, excretion and body tissues following the administration of a single dose of radioiodine.

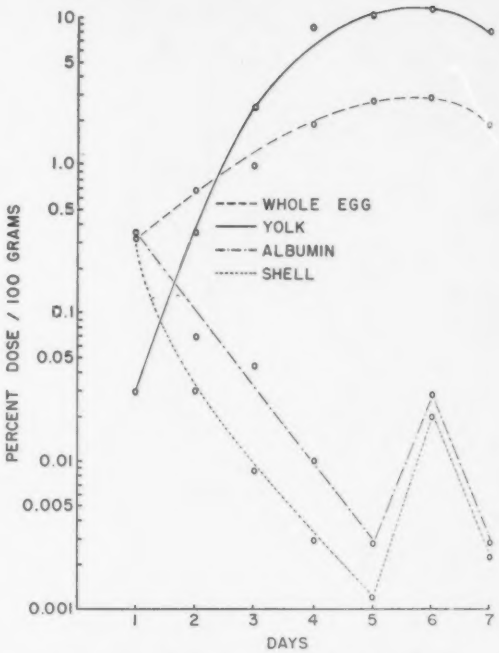


FIG. 5. The distribution of I^{131} in the different fractions of the egg after administration of a single dose of I^{131}.

Most interesting is the observation that the radioiodine content of the eggs reached a peak on the sixth day after the start of the experiment. When the eggs were separated into yolk, albumin or shell (Fig. 5) it was seen that this was due almost entirely to the yolk since both the albumin and shell showed a peak incorporation of activity on the first day after dosing and decreased rapidly after that.

It should be noted that the time for the maximum value to be established in the eggs coincided with the establishment of a plateau level in the multiple dosing experiment. However, the sum of the activity appearing in the eggs for these 6 days, after the single dose of I^{131}, was only half of the equilibrium level established with daily doses. Theoretically, these should have been equal. The reasons for the difference are not clear at this time; however, as with dairy cattle[4] it does not seem possible to use single-dose experiments to predict the levels to be attained in the eggs after long-term ingestion of I^{131}.

DISCUSSION

The handling of the chickens used for this study represents management conditions that would be duplicated in many commercial egg producing areas of the United States. The data show that a significant fraction of a dose or doses of I^{131} ingested by the laying hen could find its way, via the egg, into the human food chain. As has been demonstrated with stable iodine[2] the major portion of the I^{131} becomes located in the yolk. It is interesting to note that 90 per cent of the ingested radioactivity was excreted within 24 hr. Of that remaining in the body, most was accounted for as I^{131} incorporated into ova that are in the process of depositing yolk material prior to being formed into eggs. Little of the I^{131} was located in the edible fleshy portions of the bird.

The time necessary for the yolk to exhibit its maximum content of I^{131} was 6 days in both the single and multiple dose experiments (Figs. 2 and 5). Thus, the hazard from I^{131} would be more critical several days after the hens were briefly exposed to I^{131} rather than immediately. In contrast, the milk of cows given a single dose of I^{131} could be expected to show maximum levels in a few hours and then decline rapidly.[3]

Other radioisotopes of iodine as well as I^{131} are produced in large amounts during nuclear fission. Most of these, however, have short half-lives which, hazardwise, compensates for their relatively abundant production. Of these, I^{133} is the most significant since it has a relatively long half-life of 22 hr. Using Fig. 4 one would estimate that the short half-life would prevent I^{133} and the other short-lived radioiodines, from being as pressing a problem as I^{131} under both single and multiple exposure conditions.

Also, the data of Fig. 4 have significance in showing that where laying hens were ingesting I^{131} over a period long enough to allow the establishment of an equilibrium level of I^{131} in the eggs, complete removal of the contaminating I^{131} of the food or successful remedial measures could be expected to show significant declines in egg yolk I^{131} only after a lag period of about a week. This is unlike milk where the decline is quite rapid.[4]

It should be kept in mind that the albumin of egg was quite unlike the yolk in its reaction and that it appeared to reflect the levels of I^{131} in the body more than did the yolk. The albumin, along with the flesh of the hen, would be a safer source of food for the human population after a nuclear incident than would the egg yolk.

A question yet to be answered is the likelihood of contamination of the food supply intended for feeding to poultry. One would suspect that unless inhalation was a major pathway of uptake, only poultry allowed access to ranges or exercise yards would show significant levels in their eggs. Thus evaluation of the hazard likely to be encountered would depend on the agricultural practices typical of the immediate area. In the United States, where most hens are confined and fed commercial grain mixes that have undergone considerable storage since harvesting, the likelihood of contamination is low. Also, additional protection is afforded the human population by the delay encountered in collection, transportation, grading and storing that eggs usually encounter on the way to the consumer. In some areas, where small flocks of chickens are kept, it is customary to allow chickens access to the outdoors. Such a situation could result in high relative amounts of I^{131} being found in the eggs.

SUMMARY

(1) Iodide I^{131} was administered orally to laying hens daily for a period of 19 days. Steady state occurred at about 7 days, with 8 to 9 per cent of the daily dose being found in the eggs each day from the seventh through the nineteenth days.

(2) Approximately 90 per cent of the daily dose was lost from the body each day. The amount remaining in the body after 19 days of daily intake was equivalent to 70 per cent of the daily dose.

(3) Of the radioactivity of the egg, the major portion was contained in the yolk, which required 7 days to come to a plateau value. The albumin and shell contained little radioactivity and equilibrated with the dietary I^{131} within a day or so.

(4) Intravenous injection of I^{131} gave the same distribution pattern as oral I^{131}. After a single dose only 10 per cent was left in the body at 24 hr. In the next 6 days the remaining activity was lost mostly through the eggs, very little appearing in the excreta.

(5) After a single dose of I^{131} the maximum value in eggs was observed on the sixth day after dosing with the time pattern being governed by accumulation in the yolk.

REFERENCES

1. *Radioactive Materials in Food and Agriculture*, FAO Atomic Energy Series, No. 2, P. 115, Rome (1960).
2. J. Straub, *Debrenceni Szemle* **6,** 129 (1932).
3. F. W. Lengemann, E. W. Swanson and R. A. Monroe, *J. Dairy Sci.* **40,** 387 (1957).
4. F. W. Lengemann and E. W. Swanson, *J. Dairy Sci.* **40,** 216 (1957).

Health Physics Pergamon Press 1961. Vol. 6, pp. 32–35. Printed in Northern Ireland

LEAKS IN STANDARD SEALED TELETHERAPY SOURCES*

R. J. CLOUTIER and MARSHALL BRUCER
Medical Division, Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tennessee

(*Received* 21 *September* 1960)

Abstract—Teletherapy sources encapsulated in the United States and Canada from 1954 to 1959 were generally of the standard, single capsule, sealed type. One feature of the standard capsule was believed to be its tight seal and its ability to maintain this integrity. Late in 1958, a standard capsule source at the Oak Ridge Institute of Nuclear Studies (encapsulated $1\frac{1}{2}$ years earlier with cobalt-60 metal pellets) developed a leak. This leak resulted in from 30 to 100 mc of activity being released before it was detected. Subsequent investigation of other sealed sources in the United States revealed that thirteen of 200 standard teletherapy capsules had developed leaks. A review of the possible causes of leakage, and of the standard capsule design, fabrication, sealing and initial and subsequent leak testing, has resulted in recommended changes to reduce the probability of future capsule failures. The incidence of failures already encountered, however, has made apparent the importance of routine leak testing of encapsulated sources.

INTRODUCTION

A SEALED source is generally defined as being any source that is encased in a capsule designed to prevent the leakage or escape of the radioactive material. The importance of preventing uncontrolled leakage of radioactive material was recognized in the early 1900's. Radium sources made at this time were frequently encapsulated. These capsules, however, sometimes failed, and widespread contamination often resulted with the attendant risk of persons ingesting or inhaling the radium or radon. The risk of source capsule failure is not a new problem.

When, in the late 1940's, radioisotopes became generally available, medical personnel recognized that the radiation from one of these isotopes, cobalt-60, could be used to advantage. Cobalt-60 offered a means of treating patients with photon energies in the supervoltage range. To produce photons of comparable cobalt-60 energy, two-million-volt peak X-ray units were required. These units are expensive to build and maintain.

* Under contract with the United States Atomic Energy Commission. Presented at the meeting of the Health Physics Society, 1 July 1960.

It was recognized in the early design development of teletherapy units that they must be inherently safe and that a desirable feature would be the standardization of source capsules so that sources could be interchanged between units.[1] Thus a source that through decay was no longer adequate for one program could be transferred to another teletherapy unit that did not require so high a radiation output rate. The design of the first standard cobalt-60 teletherapy source was selected by the members of the X-ray industries, Atomic Energy of Canada, Ltd., and the United States Atomic Energy Commission in November 1953.[2]

DESCRIPTION OF THE FIRST STANDARD COBALT-60 TELETHERAPY CAPSULE

Most of the cobalt-60 sources produced for teletherapy units after January 1954 were of the standard cobalt-60 type. This was a single capsule, sealed by means of lead O-rings. The capsule (Fig. 1) consisted of a cap of tungsten alloy. Tungsten alloy was chosen because of its high density, its cost, and its ability to be machined. By placing the high density tungsten close to the source, where it could be used to best advantage as shielding, the overall weight of the teletherapy unit could be reduced. The

end of the cap was open, thus permitting the γ-rays to pass through unattenuated in the direction of the useful beam. A cup of stainless steel fitted into the cap. A tungsten plug designed according to the shape of cobalt to be inserted was then fitted into the cup. Two lead rings were used to seal the unit. The first O-ring fitted at the shoulder of the plug, and the second fitted at the lid of the stainless steel can. The units normally held 1000 curies; hence, they were loaded remotely. To load the capsule the cobalt-60 was placed into the plug. The plug was then inserted into the stainless steel can, and the cap was placed over the can. With the lead O-rings in place, the units were screwed together with sufficient pressure to cause the lead O-rings to cold flow and form a leak-tight seal.

The units were then leak tested by the wipe method. This method consists of wiping the source capsule with filter paper and checking the wipe for contamination. Sources that showed negligible contamination were considered properly sealed. These sources were then held for about 30 days, when a subsequent leak test was made to ensure that the seal was maintained. If the source capsule passed both leak tests, it was shipped to the purchaser for installation in the teletherapy head.

DESCRIPTION OF REPORTED LEAKS

The design of the capsule and its testing before being used were believed in 1953 to be adequate for the prevention of leaks. In fact, routine leak testing after installation of the source was not required by the Atomic Energy Commission. The NCRP in the National Bureau of Standards Handbook 54, *Protection Against Radiations from Radium, Cobalt-60, and Cesium-137*, did, however, mention that smear testing of the source should be carried out.

A source, which was encapsulated in a standard capsule and had passed the 1957 standard wipe leak test, was placed in an Oak Ridge Institute of Nuclear Studies teletherapy unit. About a year and a half later, a leak developed and moderate contamination resulted. The capsule contained about 650 c of cobalt-60 in the form of cobalt metal pellets.

The manner in which the ORINS leak was detected is of interest, for some of the leaks noted at other installations were detected in a similar manner. A routine smear contamination check of the medical facilities had indicated that slight contamination existed. While a recheck of the areas was being carried out to determine the source of the contamination, a technician who had been working in the teletherapy rooms approached a NaI detector used to scan patients. The scaling unit attached to the detector began to indicate the presence of radioactive material. When the technician retreated and reapproached the detector, it was immediately apparent that he was contaminated. A spectrum of the contaminant indicated that it was cobalt-60. Monitoring of the teletherapy areas, where the technician worked, showed that contamination was more widespread in these rooms. Subsequent surveying showed that the contamination was from teletherapy head no. 1 of a two-headed teletherapy unit. The estimate made after an analysis of the many surveys and analysis of smears taken in the teletherapy area and in the cone of the teletherapy head indicated that from 30 to 100 mc of cobalt-60 had been released. Contamination of the medical facilities, although extensive, was not too difficult to decontaminate and only standard cleaning methods were required.[3]* The early detection of the leak and the control procedures used for the subsequent decontamination of the area were sufficient to prevent personnel exposures in excess of one-tenth of the permissible external and internal radiation exposures.[3] The teletherapy head containing the leaking capsule was sealed in polyethelene and the head was removed from its yoke. The head was sent to the Oak Ridge National Laboratory where, in a hot cell, the source was removed and examined. There was no external, visible sign of damage to the capsule. The source was removed from the capsule and the capsule and seal were examined. Some damage to the lead seal was noted.

Since one leak had occurred with the standard capsule, it was deemed advisable to conduct a pilot study and get in touch with some owners

* Reference (3) gives a detailed description of the leak and the problem of decontamination.

of this type of source capsule and have them leak test their unit. At this time there were about 200 of the old O-ring standard capsules in use. Most of these sources were made by the Oak Ridge National Laboratory and the Canadian Chalk River Laboratories. Fifty owners of the old capsule type of sources were requested to leak test their units. The results were favorable in that there were no other apparent leaks. The leak testing technique used was similar to that recommended in NBS Handbook 54.[4] This method consists of smearing with a piece of filter paper either the capsule or parts of the unit that come into proximity with the capsule during some part of its movement.[3–5]

Since the request for leak testing was made, thirteen of the 200 standard capsules have developed leaks. Incidentally, although nonstandard capsules are not included in this report, it is known that three capsules of nonstandard design have developed leaks.

Decontamination costs for these leaks were always high. The costs, however, depended greatly on how soon the leak was detected after it started, the size of the leak, and the nature of the room in which the leak occurred.

CAUSE OF THE LEAKS

Although it is impossible to give the exact cause of the leaks, because complete studies were not carried out, causes can be postulated. One cause could be that the seals initially were not complete and that the test method used initially was inadequate. The method, as mentioned earlier, was the wipe test. This method is not a critical one because it does not actually measure the integrity of the seal.

For all leaks reported to date, there was a delay of more than one year between the encapsulation and the time that the leak was noted. This may be attributed to the fact that the source is a metal and many of them are coated with gold or nickel to prevent the surface from oxidizing. It is believed that cobalt oxide is the material that leaked from the capsules; if so, it would have been restricted in its formation and release. Therefore, a period of time would have to elapse before sufficient cobalt oxide was available for release.

Another possibility is that the seal was good at the time that the source capsule was checked, and that it failed only later. Seal failure could have been brought about by two causes. One, the failure could have resulted from the mechanical movement of the capsule or by the movement of the source itself inside the capsule. In general, the source is prevented from moving by means of shims. Repeated movement of the source could possibly have caused the failure of the mechanical seal, for lead acts like plastic and does not resume its original shape if deformed. The second possibility is that the heat generated by the source could have caused the seal to fail by causing expansion of the components, or could have melted the O-ring seal and caused it to flow. On cooling it would not return to its original shape. A kilocurie of cobalt-60 under certain conditions could reach a temperature high enough to cause this to occur. In most teletherapy units, however, there is a large heat sink in the form of the shielding and the shutter wheel to carry off the heat and prevent excessive temperatures from developing. In the loading, transfer, or storage of the source before loading, there is not always such a desirable heat transfer medium and high temperatures could be attained. The lead O-ring of the ORINS source that failed was damaged in one place. The damage was similar to that expected from melting. If failure was due to melting, it is not believed that the melting could have taken place in the teletherapy head because of the vast heat sink available to the source. In storage before loading, however, there were times when the source was in air with limited heat transfer. The O-ring failure may have occurred at this time. This, however, does not explain why the leak developed many months after the source was placed in the unit.

DESCRIPTION OF THE NEW STANDARD CAPSULE

Because the difficulty appeared to be at the O-ring seal, it was realized that the old standard capsule was not adequate, and that a new type seal was required. The size of the capsule was retained in order to permit the continued interchange of the units. The new capsule,

FIG. 1. Old standard teletherapy capsule.

FIG. 2. New standard teletherapy capsule.

recommended by the Fifth X-ray Industrial Conference in December 1958[3] is shown in Fig. 2. The capsule consists of a stainless steel can into which the cobalt is placed. The can is closed with a Heliarc-welded cap.[6] The sealed can is then placed in another stainless steel cup. A cap is placed on this cup and the unit is Heliarc welded closed. The source is now encapsulated in a double can, both of which are welded closed. As a check on the integrity of the seal, the capsule is leak tested by the heated water or helium leak test method before being installed in a teletherapy unit. The leak test methods now used actually measure the integrity of the seal. A wipe test, however, is still performed to determine the degree of contamination of the outside container.

The capsule after passing the leak and wipe test is placed in a capsule holder that has the same outside physical dimensions as the old standard capsule. Most of the units now being made are of this type. To date no failures of the new capsule have been reported.

SUMMARY

It is now known that thirteen cobalt-60 teletherapy capsules, encapsulated in the standard O-ring type teletherapy capsule have developed leaks. About 200 of these capsules are still in service. It is impossible to state the cause of the capsule failures that have already occurred, or to state that all the capsules that are going to fail have already done so. Certain general things can be said. Capsules that have leaked were in service for more than a year before leaks occurred. Also the cobalt dust that leaked usually resulted in rather widespread contamination before detection.

Until recently, AEC regulations did not require the routine checking of cobalt teletherapy capsules for leakage; however, the AEC now requires leak testing every 6 months. The new teletherapy capsule has been designed to reduce the possibility of leakage; however, it must be remembered that during the initial design work of the old capsule it was believed that the initial capsule "wouldn't leak in a million years." The problem of ensuring the integrity of capsules should be re-examined at each site, and especially at sites using the old type of capsule. Although this paper has dealt only with teletherapy sources, a comparable number of leaks has been noted for other uses of sealed sources. Periodic leak testing should be carried out regardless of the type of capsule or its use, if the problems of contamination and the attendant high cost of decontamination are to be avoided.

REFERENCES

1. M. Brucer, *Brit. J. Radiol.* **27,** 410 (1954).
2. M. Brucer *et al.*, *Teletherapy Design Problems III.* United States Atomic Energy Commission Report ORO-127 (1954).
3. M. Brucer and E. B. Anderson (Editors), *Radiation Protection at the Medical Division.* United States Atomic Energy Commission Report ORINS-37. In preparation. Draft copies available from the Medical Division.
4. National Bureau of Standards Handbook 54, *Protection Against Radiations from Radium, Cobalt-60, and Cesium-137.* 1 September (1954).
5. National Bureau of Standards Handbook 73, *Protection Against Radiation from Sealed Gamma Sources.* 27 July (1960).
6. E. E. Pierce, *Remote Welding of Stainless Steel Containers.* ORNL 2280, 22 April (1957).

Health Physics Pergamon Press 1961. Vol. 6, pp. 36–40. Printed in Northern Ireland

EFFECT OF A CARBONIC ANHYDRASE INHIBITOR (DIAMOX) ON Cs^{137} EXCRETION BY RATS*

C. R. RICHMOND and J. E. FURCHNER
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico

(*Received* 20 *February* 1961)

Abstract—Oral administration of Diamox at levels of 6.5, 12.8 and 27.0 mg/kg per day for 20 days elicited similar responses in accelerating the excretion of Cs^{137} administered intraperitoneally to rats. The Cs^{137} retention functions for treated and untreated groups could be described by the sum of three exponential components. Analysis of variance showed significant differences between the control and treated groups. Differences among the treated groups were not significant.

Diamox treatment increased the turnover rates of the two components normally having the largest rate constants. The net effect of enhanced excretion was a 20 per cent reduction in the radiation dose received by the Diamox-treated animals as compared with control animals.

INTRODUCTION

2-ACETYLAMINO-1:3:4-THIADIAZOLE-5-SULFONAMIDE (Diamox) is a diuretic and acid base regulator of low toxicity, which acts specifically to inhibit carbonic anhydrase. Diuresis results from the effect of the drug on the reversible reaction $H_2O + CO_2 \leftrightarrows H_2CO_3$ occurring in the kidneys. The net result is a renal loss of HCO_3^- ion and subsequent increased loss of water, sodium and potassium. One might reasonably expect this compound to accelerate the excretion of other monovalent cations such as rubidium and cesium. Metabolic similarities among rubidium, cesium and potassium have long been recognized.[1–5] The purpose of this study was to investigate the effect of Diamox on the excretion of Cs^{137} by rats.

Cesium-137 is a high yield product of uranium and plutonium fission, which is readily absorbed from the intestinal tract and deposited predominantly in muscle tissue. One must consider the potential hazard from radiocesium as a by-product of large-scale power production and other peaceful uses of nuclear energy.

* Work performed under the auspices of the U.S. Atomic Energy Commission.

METHODS AND MATERIALS

Twenty-four adult male Sprague–Dawley rats were used. These animals were 64 days old at the time of the experiment and weighed 273.9 ± 7.1 g ($\bar{X} \pm \sigma$). Each animal was injected intraperitoneally with 0.75 μc of carrier-free Cs^{137} as the chloride in 0.1 ml of solution adjusted to a pH of 5. At 30 min following administration, the animals were divided into four equal groups and the Cs^{137} activity in each animal determined by whole body counting with a 4π liquid scintillation detector.[6] Activity measurements were made at subsequent intervals over a 76-day period.

Immediately after administration of the Cs^{137}, all animals were placed on a dietary regimen which consisted of Purina chow and tap water *ad libitum*. Diamox was added to the drinking water of the three experimental groups to give concentrations of 0.045, 0.090, and 0.180 mg/ml, respectively. Diamox treatment was continued for a period of 20 days. The average daily Diamox intake per kg body weight was calculated from the average weight of the animals and their daily fluid intake. Daily intake of Diamox by the three experimental groups was 6.5, 12.8 and 27.0 mg/kg,

respectively (Table 1). Increase in body weight was used as an indication of the physiological condition of the animals during the experiment. No observable differences were found between control and treated groups.

At completion of the experiment, the Cs^{137} whole body retention data were coded for analysis by a 704 computer, which was programmed to perform iterative least square calculations. Coding procedures called for retention values (Y) at each time (t) and the estimated intercept and rate parameters of the retention function. Each value of Y was weighted by a factor proportional to $1/Y$. The 704 procedure avoids the subjectivity associated with eye-fitting a line to a number of experimental points and, by iteration, assures that best fit criteria are equally applicable to all components simultaneously. Preliminary graphic analysis of the retention data suggested the presence of three exponential components.

The integral of each retention function from t_0 to t_∞ was calculated and used for assessing the effect of Diamox intake levels on Cs^{137} retention. Total area under the retention function was selected as it is proportional to the total number of atoms which disintegrate within the body and, therefore, is proportional to the radiation dose. The contribution from physical decay of Cs^{137} to the retention function is insignificant because of its long half-life (about 28 years).

RESULTS

Table 1 gives the average whole body retention values for each group of six rats at each time of measurement. In each case, whole body retention between 0 and 76 days could be described mathematically by the sum

Table 1. *Average whole body retention (per cent) of* Cs^{137} *as a function of time in control and Diamox-treated animals*

Day	Group*							
	A	σ^+	*B*	σ	*C*	σ	*D*	σ
0	100.00		100.00		100.00		100.00	
1	84.70	2.06	75.90	8.87	79.95	5.15	81.04	3.04
2	75.91	1.64	67.28	7.89	69.88	5.15	70.71	3.41
3	68.60	2.22	59.89	6.96	62.85	5.00	61.71	4.04
6	54.50	2.99	46.36	5.24	48.40	5.08	47.57	3.86
7	51.09	2.62	43.26	4.47	44.85	4.78	43.53	3.68
10	41.32	2.42	34.52	3.46	35.34	4.03	33.86	3.87
13	34.32	2.24	27.19	2.23	28.47	3.35	26.13	3.37
15	30.78	2.37	23.67	1.83	24.50	2.92	23.10	3.19
20	22.26	1.47	16.18	1.12	16.55	2.15	14.93	2.69
23	18.16	1.34	13.14	1.04	13.48	1.84	12.08	2.53
28	13.41	1.08	9.85	0.75	10.05	1.50	8.74	2.20
34	9.63	0.77	7.15	0.73	7.07	1.10	6.07	1.86
38	8.14	0.69	6.19	0.75	5.96	1.02	4.87	1.70
44	5.50	0.52	4.22	0.19	4.22	0.69	3.23	1.28
52	3.54	0.41	2.79	0.49	2.67	0.55	2.05	0.95
58	2.56	0.32	2.06	0.40	1.93	0.42	1.46	0.77
69	1.41	0.22	1.19	0.28	1.10	0.27	0.77	0.49
76	1.01	0.17	0.81	0.27	0.70	0.19	0.48	0.33

* Groups *A*, *B*, *C* and *D* (six animals each) correspond to daily intakes of 0, 6.5, 12.8 and 27.0 mg Diamox/kg, respectively.

$$\sigma^+ = \pm\sqrt{\left[\frac{\Sigma(X-\bar{X})^2}{n-1}\right]}.$$

of three exponential components. At 76 days after administration, the retention of Cs^{137} had decreased by a factor of 100 in the control group. Long-term experiments with rats at this laboratory indicate that additional exponential components are not needed to describe Cs^{137} retention functions out to 140 days, at which time the original burden had decreased by a factor of 10^4.[6]

Table 2 gives the machine calculated values for the intercept and rate parameters that describe each exponential component of the total retention curve for each group. The area under each exponential portion is given and is expressed also as a percentage of the area under the total retention function. Total area under the retention functions for groups receiving Diamox treatment averaged ~80 per cent of that for animals receiving no Diamox.

DISCUSSION

An analysis of variance using retention values for all animals of each group at each time of measurement showed that there were significant differences among all the groups at the 5 per cent level. DUNCAN's test[7] was then applied and indicated that the controls differed significantly from each of the treated groups but that no significant differences existed among the treated groups. Additional tests were performed to test for significant differences (5 per cent level) between the average retention values for each group and those for all other groups at each time of measurement. On days 1 and 2, group *A* differed only from group *B*. Group *A* differed only from group *D* on days 58 and 69. On day 76, group *A* differed from groups *C* and *D*. At all other times, group *A* differed from each of the treated groups. There were no differences among the treated groups except between groups *B* and *D* on day 76. These data indicate that the various Diamox treatment levels elicited similar responses among the treated groups.

The most noticeable effects of Diamox treatment on the total retention pattern are reflected in the intercept values of the third exponential portion and in the values of the rate constants of the first and second components. The rate constant of the third component of the retention functions for all four groups was essentially the same. Cesium-137 atoms associated with the first and second components apparently are excreted at an increased rate from the animals receiving Diamox. This leaves a smaller fraction of the administered dose available for deposition at sites which lose Cs^{137} atoms at the slowest rate. Assuming each component of the retention curve represents a body compartment, the net effect of Diamox treatment is an increased rate of loss from the compartments which normally have fastest and intermediate turnover rates. Rate of loss of Cs^{137} bound in the compartment with the slowest turnover rate is not increased by Diamox, indicating that elimination from this compartment is not governed by renal function. However, as a consequence of accelerated loss from the other two compartments, relatively less Cs^{137} is bound in the compartment with the smallest rate constant. This is of practical significance with regard to radiation exposure, as the major fraction of the radiation dose is related to the compartment with the slowest turnover time.

Because of the direct proportionality between radiation dose and the total areas given in Table 2, the total dose delivered to the Diamox-treated animals would be about 20 per cent less than the dose received by the nontreated group.

The technique of comparing areas under the whole body retention functions to assess the effect of various treatments has several distinct advantages over the commonly used techniques in which the activities excreted by the different groups are compared at several days or a week after administration. Examination of Table 1 shows that the difference in Cs^{137} retention between the control and treated groups varies with time after administration from ~9 per cent on day 3 to ~34 per cent on day 76. Thus a single observation of the difference between control and treated groups would yield relatively little information. Use of the 4π whole body counter allows one to make repeated precise measurements of the activity in the same animals following administration of extremely small doses of γ-emitting radionuclides. This technique can be extended to some β-emitting radionuclides by using similar, but

Table 2. Parameters of Cs^{137} retention functions for control and Diamox-treated rats

Diamox (mg/kg per day)	Component	a* (%)	k* (fraction per day)	T_b† (days)	Area‡	
					(% days)	(% of total)
0	1	17.34	0.75918	0.91	22.7	1.7
	2	32.38	0.10094	6.87	320.8	24.5
	3	50.19	0.05181	13.38	968.7	73.8
	Total				1312.2	
6.5	1	21.11	1.66093	0.42	12.7	1.2
	2	46.97	0.12042	5.75	390.1	36.4
	3	31.90	0.04759	14.56	670.3	62.5
	Total				1073.1	
12.8	1	18.18	1.27353	0.54	14.3	1.4
	2	46.50	0.12110	5.72	384.0	36.7
	3	35.33	0.05459	13.73	647.1	61.9
	Total				1045.4	
27.0	1	18.30	1.02347	0.68	17.9	1.8
	2	48.03	0.12240	5.66	392.4	38.3
	3	33.71	0.05487	12.63	614.4	59.9
	Total				1024.7	

* From $R_t = \sum_i^n [a_i \exp(-k_i t)]$, where R_t is the retention at any time t, a_i and k_i are the intercept and rate constants, respectively, and t is days.

† $T_b = \dfrac{0.693}{k}$.

‡ Total area is obtained by integrating the retention function from t_0 to t_∞.

more sensitive, counters which are capable of detecting bremsstrahlung.

Excretory patterns of radiocesium in rats have been shown by MRAZ and others to be influenced by natural foodstuffs in the diet.[8] After assimilation by the body, however, organic compounds such as parathyroid extract, cortisone, and dinestrol diacetate show little effect on altering the excretory pattern.[9] Alterations in dietary potassium levels, as one might reasonably expect, appear to accelerate Cs^{137} excretion.[10] Results of this investigation indicate that Diamox also enhances to a limited degree the excretion of Cs^{137}. This finding is consistent with a large body of information which shows that potassium and cesium appear to be physiological analogs.

SUMMARY

Oral administration of Diamox at levels of 6.5, 12.8, and 27.0 mg/kg per day for 20 days elicited similar responses in accelerating the excretion of Cs^{137} administered intraperitoneally to rats. A 4π liquid scintillation counter was used to determine changes in Cs^{137} activity in the live animals for a period of 76 days. The whole body retention functions for treated and untreated animals were then reduced into three exponential components by a 704 computer. Diamox treatment increased the turnover rates

of the two components normally having the largest rate constants. The net effect of the enhanced excretion was a reduction of ~20 per cent in the radiation dose received by the Diamox-treated animals as compared with non-treated animals.

Acknowledgments—The authors acknowledge the valuable counsel and assistance of W. LESTOURGEON (Group T-1) in deriving the parameters of the retention functions and in performing statistical tests. Technical assistance of Miss G. A. TRAFTON (Group H-4) is also acknowledged.

REFERENCES

1. S. RINGER, *J. Physiol.* **4,** 370 (1882).
2. R. H. FOLLIS, *Amer. J. Physiol.* **138,** 246 (1943).
3. R. A. MACLEOD and E. E. SNELL, *J. Biol. Chem.* **176,** 39 (1948).
4. A. S. RELMAN, *Yale J. Biol. Med.* **29,** 248 (1956).
5. A. S. RELMAN, A. T. LAMBIE, B. A. BURROWS and A. M. ROY, *J. Clin. Invest.* **36,** 1249 (1957).
6. C. R. RICHMOND, Los Alamos Scientific Laboratory Report LA-2207 (1958).
7. D. B. DUNCAN, *Biometrics* **11,** 1 (1955).
8. F. R. MRAZ and H. PATRICK, *J. Nutrition* **61,** 535 (1957).
9. F. R. MRAZ, M. LENOIR, J. PINAJIAN and H. PATRICK, *Arch. Biochem. Biophys.* **63,** 73 (1956).
10. F. R. MRAZ and H. PATRICK, *Proc. Soc. Exp. Biol., N.Y.* **96,** 497 (1957).

Health Physics Pergamon Press 1961. Vol. 6, pp. 41–45. Printed in Northern Ireland

A SIMULATOR FOR γ-RAY EXPOSURE FROM NUCLEAR FALLOUT*

D. G. BAKER, J. C. F. MacDONALD† and C. G. HUNTER‡
Banting and Best Department of Medical Research, University of Toronto, Toronto 5, Canada

(*Received* 9 *November* 1960)

Abstract—A device is described by which laboratory animals may be irradiated at dose rates which are varying in a way similar to those from the γ-radiation from radioactive fallout. The animals are moved relative to a fixed source of cobalt-60 in such a way that the dose-rate to which the animals are exposed varies as a power function of time. Experimental verification of the theory of the device is presented. A method of achieving a range of total exposure doses in a particular time interval is described.

The $LD_{50/30}$ for mice exposed to simulated fallout of 96 hr duration was 1109 r compared to 1008 r when the dose was delivered at a constant dose rate over the same time interval, or 553 r when it was delivered at 65/min using a 250 kV X-ray source.

INTRODUCTION

ALTHOUGH total dose is recognized as the most important factor in the production of the physiological response to ionizing radiation, the significance of dose rate and over-all time of irradiation is also well established. The effects of single, intermittent, and protracted exposures to ionizing radiation have been studied in a wide range of biological material. In general, fractionating the dose or reducing the dose rate has been shown to decrease the severity of the immediate reaction.[1–3]

Observations made in test areas after the detonation of nuclear devices indicate that the exposure dose rate due to the γ-radiation from early fallout varies as a power function of time.[4] In view of the importance of dose rate in dose-response relationships it is of interest to study the response of laboratory mammals to this kind of irradiation, and to compare it with the response under conditions of constant dose rate.

This report describes the construction of a device which makes possible the irradiation of small laboratory mammals at dose rates which are varying in this way. The desired variation of exposure dose rate is obtained by altering the distance between the target material and a fixed point-source of approximately 45 c of cobalt-60. This distance is slowly increased in such a way as to transform the inverse-square-law dependence into the required power function of time. The source activity is considered to be constant during the period of each experiment.

DERIVATION OF THE GENERAL EQUATIONS

The exposure dose rate at a point within the fallout zone will be expected to increase with time as the activity of fallout material in its immediate vicinity increases. A peak dose rate will be reached when the effects of decay and the rate of deposition just balance. A short time after this, the variation of dose rate with time has been observed[4] to follow the empirical relationship:

$$R_t \propto t^{-m} \qquad (1)$$

where R_t is the exposure dose rate in roentgens per hour at the point of interest, t is the time in hours after detonation, and m is a constant

* Work supported by a Grant DRB 9910-20 and 9310/27 from the Defence Research Board of Canada

† Present address: Cancer Clinic, Victoria Hospital, London, Ontario, Canada.

‡ Present address: Shell Research Laboratories, Finsbury Circus, London, England.

determined principally by the distribution of fission products species. If this relation is considered valid between an initial time t_i and final time t_f, the dose rate at any time $t_i < t < t_f$ is given by:

$$R_t = R_{t_i}\left(\frac{t_i}{t}\right)^m \text{ r/hr} \tag{2}$$

The variation of exposure dose-rate R_x with distance x cm from a long-lived point source of activity M mc and specific γ-ray emission K cm^2 r mc^{-1} hr^{-1} is given by:

$$R_x = \frac{MK}{x^2} \text{ r/hr} \tag{3}$$

The combination of equations (2) and (3) by the requirement that $R_t = R_x$ leads to the general equation for the motion of the target:

$$x = \left(\frac{t}{t_i}\right)^{m/2} x_i \text{ cm} \tag{4}$$

where the subscript i refers to the initial condition. If the subscript f refers to the conditions at the end of the experiment then the distance traveled by the target during the experiment is given by:

$$x = (x_f - x_i) = x_i\left[\left(\frac{t_f}{t_i}\right)^{m/2} - 1\right] \text{ cm} \tag{5}$$

The integrated exposure dose at the target during the time $\Delta t = (t_f - t_i)$ of the experiment is:

$$E_{if} = \frac{R_{t_i}\, t_i}{1 - m}\left[\left(\frac{t_f}{t_i}\right)^{1-m} - 1\right] \text{ r} \tag{6}$$

This expression can be evaluated for all values of $m \neq 1$. For the particular case $m = 1$:

$$E_{if} = R_{t_i} \cdot t_i \cdot \ln\left(\frac{t_f}{t_i}\right) \text{ r} \tag{7}$$

For a chosen set of values of t_i and t_f, it is to be noted that the integrated exposure dose E_{if}, the final distance x_f, and the distance traveled by the carriage x, all depend upon the choice of x_i.

DESCRIPTION OF THE DEVICE

Fig. 1 shows the arrangement of the fallout device. The 45-c cobalt-60 source is contained in a small capsule which can be exposed by moving it from a storage castle to a predetermined position using controls outside the protective walls. A carriage, made of light metal and plywood and supporting plastic animal cages, runs freely along two sets of tracks in a direction radially outward from the source. The movement of the carriage is constrained by the rotation of a motor driven shaft whose radius varies along its length in a regular manner, as depicted schematically in Fig. 2. The shaft is enscribed with a closely spaced helical groove from which a cord unwinds as the shaft turns. A falling weight pulls the cages away from the source against this constraint. The shaft turns at a constant rate, but since the radius changes along its length, the length of cord unwound decreases with each successive revolution. In this way the exposure dose rate is made to vary as the power function of equation (1).

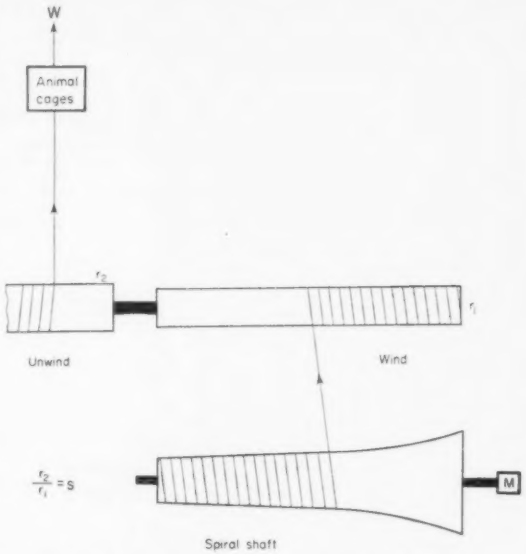


FIG. 2. Diagram for control mechanism of fallout device.

In the design of the tapered shaft, the assumption is made that the radius at any point along the shaft is great compared to the spacing between adjacent turns. On this assumption the length of the cord unwound per revolution is just equal to the circumference of

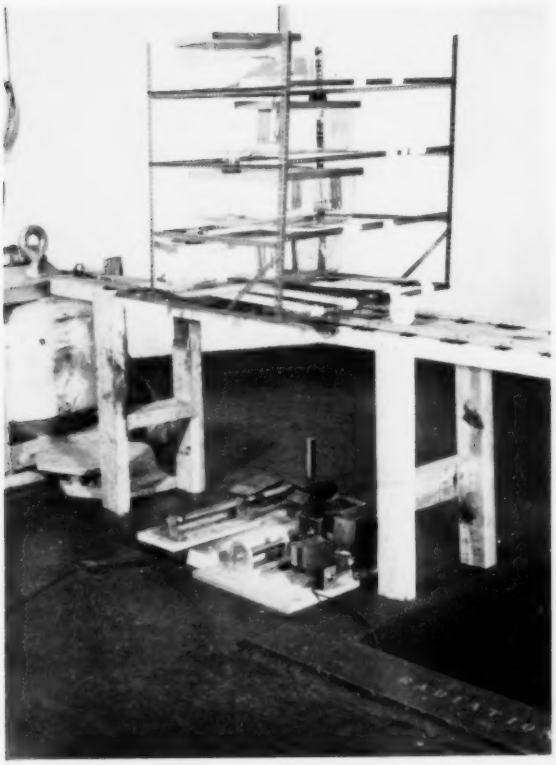


FIG. 1. Arrangement of the fallout device.

BLANK
PAGE

the shaft at that point. The radius of the shaft at the position of the nth turn can be readily calculated by:

$$r_n = \frac{x_i(t_i)^{-m/2}}{2\pi}\left[(t_n)^{m/2} - (t_{n-1})^{m/2}\right] \quad (8)$$

where t_n is the time to the end of the nth turn. It is seen that the spool dimensions depend upon the choice of x_i and t_i and the exponent m. The shaft was designed on the basis of $x_i = 100$ cm, $m = 1.2$,[4] $t_i = 10$ hr and $t_f = 106$ hr after "detonation". Thus the time interval of the experiment $\Delta t = 96$ hr. Since the drive motor turns at a speed of one revolution every 2 hr, there are forty-eight turns on the shaft.

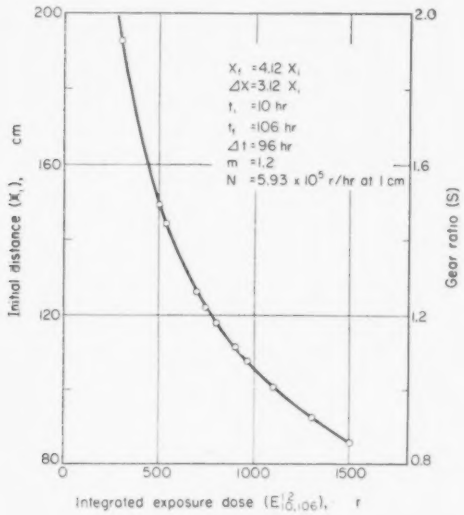


FIG. 3. Exposure dose (E).

The radius of each turn was evaluated from equation (8), and the shaft was constructed accordingly. The total length of the helical groove, and hence the total distance traveled by the target in Δt is given by:

$$\Delta x = \sum_{n=1}^{48}(2\pi r_n) = 312 \text{ cm} \quad (9)$$

These choices of the parameters result in an accumulated total dose during the exposure period which depends solely upon the activity of the source. This limitation is undesirable since a range of total doses is required. It is possible to achieve a range of total doses by the use of different values for the initial distance x_i and by the insertion of a ratio device (Fig. 2) between the tapered shaft and the target cages. By this means the distance moved by the target in the 96-hr period can be varied, even though the tapered shaft still unwinds a length of cord equal to Δx. It is found that the total exposure dose E_{if}, the initial distance x_i, the distance moved by the target ΔX, and the ratio of the radii of the intervening rollers S are all interrelated. This is illustrated in Fig. 3. A particular choice of E_{if} determines the value of the initial distance x_i, which in turn determines ΔX. The turns ratio S is then given by:

$$S = \frac{\Delta X}{\Delta x} = \frac{x_i}{100} \quad (10)$$

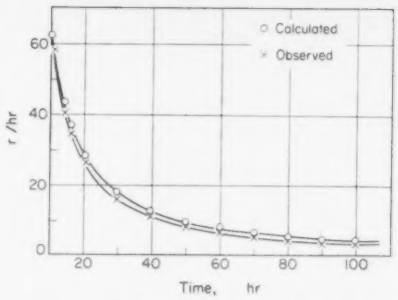


FIG. 4. Dose rate as a function of time.

TEST

The results of tests of the device are illustrated in Figs. 4–6. In Fig. 4 is shown the calculated and measured exposure dose rate as a function of time from the beginning of the experiment, under the conditions $x_i = 100$ cm and $m = 1.2$. The exposure dose rates were measured using a Victoreen "Radacon" dose-rate meter with a specially constructed

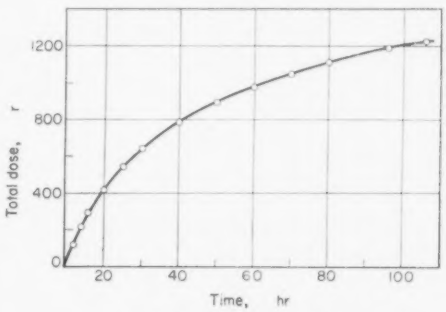


FIG. 5. Integrated dose.

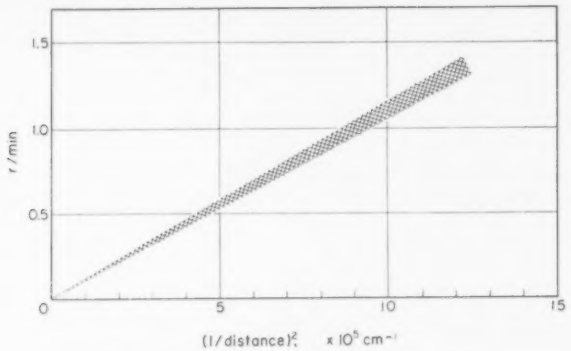


FIG. 6. Extreme variation of dose rate.

cobalt-60 thimble chamber. The variation of integrated exposure dose with time under the same conditions is shown in Fig. 5. Since the principal biological target is the small laboratory mammal, the range of exposure dose rates within plastic animal cages mounted on the target carriage has been determined throughout a typical experiment. Some of these cages are of necessity slightly to one side of the axis of travel, so the variation of dose rate from cage to cage is greatest at the shorter distances as illustrated in Fig. 6. These investigations indicated that the extreme variation of dose delivered to the animals is of the order of 4 per cent and is well within the statistical limits of biological response.

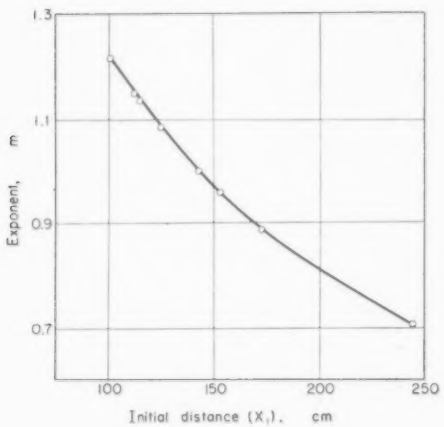


FIG. 7. The exponent (m).

In future experiments it will be of interest to determine the effects on biological response of changing the exponent m. It can be shown that by varying the initial distance x_i and by leaving out the ratio device between the tapered shaft and the target cages, the value of the exponent m may be varied over a considerable range (Fig. 7). This produces corresponding changes in the integrated exposure dose, when all the other parameters are held constant (Fig. 3).

BIOLOGICAL STUDIES

Using the "fallout" device, groups of sixty-four mice (32♂ and 32♀) were exposed to various doses of radiation delivered so as to simulate a 96-hr exposure to fallout irradiation. At the same time, comparable groups were exposed to similar total

Table 1

Statistic*		X-ray	Radiation procedure Fallout	Slow
$LD_{50/30}$	(r)	553	1109	1008
95% Confidence limits	(r)	508 to 602	1077 to 1141	975 to 1041
Slope of regression line†		−4.94	−14.2	−10.0

* All estimates of the $LD_{50/30}$ and of the regression line differ significantly from each other ($P < 0.05$).
† Regression of probit per cent survival on log dose.

doses, but delivered at a constant dose rate over the 96 hr. A third population of mice received various doses at a rate of 65 r/min, using 250 kV X-rays, (*hvl* = 1.0 mm Cu). The 30-day survival of all groups was observed. The $LD_{50/30}$ for each experimental group was determined using a probit analysis. The results are summarized in Table 1.

The data indicates that the $LD_{50/30}$ for a simulated fallout exposure of 96 hr duration is significantly greater than when the same dose is delivered at a constant dose rate over the same time interval and is twice the $LD_{50/30}$ found for X-radiation. Although the low RBE factor for Co^{60} irradiation[5] relative to the 250 kV X-rays contributes to the higher $LD_{50/30}$ value of the slow and fallout groups a significant difference still remains.

REFERENCES

1. L. H. Gray, P. Ellis, C. G. Fairchild and E. Paterson, *Brit. J. Radiol.* NS **17,** 327 (1944).
2. H. A. Blair, *Recovery from Radiation Injury in Mice and its Effect on LD_{50} for Durations of Exposure up to Several Weeks.* University of Rochester report UR-312 (1954).
3. J. B. Hursh, F. Van Slyke and G. Casarett, *Effect of Dividend Doses of X-irradiation on Survival of Rats.* University of Rochester Report UR-317 (1954).
4. S. Glasstone (Ed.), *The Effects of Nuclear Weapons.* Chapt. IX. USAEC Superintendent of Documents, Washington, D.C. (1957).
5. A. C. Upton, F. P. Conte, G. S. Hurst and W. A. Mills, *Rad. Res.* **4,** 117 (1956).

Health Physics Pergamon Press 1961. Vol. 6, pp. 46–62. Printed in Northern Ireland

THE USE OF ELEMENT-PAIRS IN RADIATION HAZARD ASSESSMENT*†

H. A. KORNBERG
Biology Laboratory, Hanford Laboratories, General Electric Company, Richland, Washington

(*Received* 28 *November* 1960)

Abstract—In using ratios of pairs of elements, such as strontium–calcium and cesium–potassium, to predict contamination levels, it is necessary to relate the ratio in the donor material to that in the acceptor material by means of a coefficient—the discrimination factor. If a constant number for any one discrimination factor relating a donor–acceptor system is used, errors can result because the discrimination factor can change with the degree of mixing of the two elements, with the environment the elements occur in as they move from one link of the biological chain to the next, and with time. Whether these changes are of practical significance in radiation hazard control, particularly where only averages are needed, is unknown. However, experimental and theoretical evidence indicates that variations in discrimination may occur under certain conditions in the field, and thereby could affect the confidence given to predictions of future contamination levels based on current measurements. This paper describes experimental and theoretical studies related to the variability of discrimination, suggesting the need for caution in using ratios, and recommends that contamination levels be reported in terms of conventional units as well as ratios.

THE application of ratios of similar elements to radiation hazard assessment has become rather general, with the strontium–calcium ratio frequently being used to predict the hazard potential of Sr^{90} in the biosphere.[2] The cesium–potassium ratio is also being employed for measuring Cs^{137} from fallout.[3] Other pairs of elements may be similarly used in the future. The purpose of this paper is to examine the bases for using ratios of similar elements to predict the hazard due to one of them as they pass through food chains and eventually to man.

RATIOS OF ELEMENT-PAIRS

Definitions

Although difficult to establish with certainty, it appears that the assumption which led to the use of the strontium–calcium pair in hazard assessment stated that the ratio of inert strontium to calcium in the human body was equal to that in the soil. It followed that a similar relationship held for Sr^{90}:[4]

$$\left(\frac{Sr^{90}}{Ca}\right)_{body} = \left(\frac{Sr^{90}}{Ca}\right)_{soil} \quad (1)$$

That this and similar expressions are not quantitatively correct was soon evident from results of many studies including extensive work by COMAR[5–8] and MACDONALD *et al.*[9] Using radioisotopes of strontium and calcium, they demonstrated that biological systems can discriminate between the two elements. As a result, equation (1) was corrected. The newer relationship is given in equation (2), where the proportionality coefficient is *DF*, the abbreviation for "discrimination factor".*

$$\left(\frac{Sr^{90}}{Ca}\right)_{acceptor} = DF\left(\frac{Sr^{90}}{Ca}\right)_{precursor} \quad (2)$$

* Work performed under Contract No. AT(45-1)-1350 between General Electric Company, Richland, Washington, and the U.S. Atomic Energy Commission.

† A brief version of this paper was presented at the Symposium on Radioisotopes on 12 October 1959[1].

* The coefficient has also been called the distribution factor[10] and "observed ratio" or *OR*[6] with *DF* being reserved for the coefficient that relates transfer of the two elements between nearly adjacent compartments, such as blood and urine. However, since truly adjacent compartments in physiological systems are difficult to define, and since the purpose of this paper is to discuss how pairs of elements are discriminated, *DF* is believed to better convey the significance of the relationship than *OR* for these purposes.

In using the relationship in Sr^{90} hazard prediction, the Sr^{90}/Ca ratio is measured in a precursor, and, from knowing the numerical value of the coefficient, *DF*, the Sr^{90}/Ca ratio in the acceptor is calculated. Results are expressed in terms of the "strontium unit" (the $\mu\mu$c of Sr^{90} per g of calcium). For any acceptor–precursor pair the *DF* itself must first be found by measuring the Sr^{90}/Ca ratios in the acceptor and the precursor.

A relationship similar to equation (2) has been used for the Cs^{137}/K ratio to predict hazards of Cs^{137}. However, no thorough attempt has been made to define the proportionality coefficient. It is usually estimated from field measurements of Cs^{137} from fallout.[11]

Justifications for using ratios

The convenience of these ratios is undeniable. As similar alkaline earth elements, strontium and calcium are readily extractable from samples being analyzed for contamination. The extract can then be used to electronically determine its Sr^{90} content and to chemically determine its calcium content. Instead of expressing the results in terms of the weight of the original material, they are expressed in terms of the weight of calcium in the original material. Further, since strontium and calcium follow similar metabolic pathways, and since the metabolism of calcium is fairly well known, relating Sr^{90} to calcium by means of the ratio appears logical.

Relating Cs^{137} to potassium is justified on a somewhat similar basis. Like strontium and calcium, cesium and potassium are in the same group in the periodic table. With suitable electronic equipment, it is relatively easy to determine the naturally occurring radioisotope of potassium, K^{40}, at practically the same time the Cs^{137} is measured. Since the ratio of K^{40} to total potassium in nature is known, Cs^{137} content of samples can be readily expressed as a Cs^{137}/K ratio. This now resembles the Sr^{90}/Ca ratio, and it suggests a parallel application of discrimination factors to describe the movement of Cs^{137} through food chains, although there is no evidence that cesium and potassium follow similar metabolic pathways.

Conditions for valid application of ratios

Because of the general application of the ratio concept in hazard assessment, it may be helpful to consider some of the conditions needed to make the application valid. The four that follow were chosen because they pertain to those applications of ratios which are capable of causing errors in predicting contamination levels based on current measurements and on the assumption that the *DF* is a constant number.

(1) When measuring ratios to find a *DF* for a given acceptor–precursor pair, it is necessary that the Sr^{90} and calcium measured in the acceptor come from the same precursor. For example, a *DF* (bone–diet) cannot be used with accuracy if, in deriving it, a significant portion of the calcium measured in the bone came from the diet before it contained the Sr^{90} being measured. This situation can be encountered when deriving *DF*s from field data.

(2) The pairs of elements should be intimately mixed and combined with similar anions. If the Sr^{90} and the calcium are not well mixed in the precursor, concentrations of the two elements measured in a sample of the precursor may not be the actual concentrations available to specific sites from which the acceptor absorbs the elements. If the two elements are not combined with similar anions, availability of the elements to the acceptor may also differ. In either case, if the pair of elements are not mixed or present as similar compounds in the precursor, any value of the *DF* measured in the laboratory under conditions of intimate mixing will probably be different under field conditions, where mixing may not have occurred. If the *DF* is measured in the field, then, for the *DF* to yield accurate predictions, the degree of mixing, or relative availabilities, of the two elements in the precursor must not change significantly with time.

(3) The *DF* should be independent of the precursor's and acceptor's environment. Plant–soil *DF*'s should be insensitive to changes in rainfall and soil types if they are to be applied to areas where such differences exist. Similarly, animal–diet *DF*'s should be insensitive to changes in dietary constituents.

(4) The *DF* should be independent of time.

An animal on a diet which contains a constant Sr^{90}/Ca ratio must contain, at any given time, a deposited Sr^{90}/Ca ratio that is related to the dietary ratio by a constant number. If this number changes with time, predictions based on a single value for the *DF* will be in error.

Before a ratio is used to predict a hazard, it would be most helpful to determine whether the four conditions are satisfied. If they are not, then, depending on the accuracy needed, it may be necessary to make allowance for the extent of deviations. Whether the use of conventional units of concentration (e.g. μc Sr^{90}/g of material in which it is found) would result in greater accuracy is not known, but it appears that there is need for caution in using ratios of elements in predicting future contamination levels. Ratios seem to lend greater confidence in predictions than is warranted, and this appears to be based on the assumption that a relevant *DF* is always measurable and is a constant. In the following, each of the four conditions listed is further examined and some of the implications are discussed.

APPLICABILITY OF DISCRIMINATION FACTORS

Origin of the element-pair

Apparently no work has been done directly to test the importance of accurately defining the precursor in determining discrimination factors. In fact, it is difficult to devise a non-subjective test for it, especially for the cesium–potassium pair, where, because of the lack of a suitable tracer for potassium, it may be impossible to prove its origin in biological materials.

Mixing of the element-pair

An erroneous *DF* can result from a difference between the quantities of the two elements *measured* in one link of a biological chain and the quantities *available* to be passed on to the next link. This error is the result of a lack of mixing of the two elements, and will not occur if both elements in the precursor link are in similar physical positions and chemical states.

Comar *et al.*[8] pointed out the importance of physical mixing of the two elements and a consequential requirement for a meaningful use of the strontium unit—that the Sr^{90}/Ca ratio measured in the soil be the same as that which is available to the plant. Another example of the lack of mixing of an element-pair is freshly fallen Sr^{90} on the surface of vegetation. In this case, any prediction of Sr^{90}/Ca ratios to be found in cow's milk based on the ratio measured in soils would be meaningless. The vegetation eaten by the cow was directly contaminated, without opportunity for the Sr^{90} to become associated with soil calcium. The importance of this was emphasized by Russell, who pointed out that the Sr^{90}/Ca ratio found in plants can be represented by a function of the sum of the Sr^{90} contributions from the atmosphere and from the soil.[12]

The mixing error can occur when the Sr^{90} in soil is expressed as a ratio with calcium, for it implies that the Sr^{90} and calcium are equally available to plants growing in the soil. To be thoroughly mixed with available calcium, the Sr^{90} which falls out has to dissolve and reach the root zone and occupy, so far as the plant is concerned, spaces equivalent to those occupied by calcium. In addition to physical movement, it has to be converted from its original chemical form in fallout to a form comparable with that of calcium in the soil.

These physical and chemical transformations undoubtedly occur. However, the time required for the transformations may be long. With fresh Sr^{90} from fallout being added to older Sr^{90} in the soil, it is reasonable to assume that the chemical and physical states of fresh and older Sr^{90} differ. The older the Sr^{90} is, the more likely it is that the Sr^{90}/Ca ratio in soil is meaningful. Unfortunately, for purposes of hazard prediction, the older Sr^{90} has physically decayed more than the fresher Sr^{90}.

Stated more explicitly, only part of the Sr^{90}/Ca ratio measured in soil is applicable to predicting the ratio that will appear in plants. Let us say that the total concentration of Sr^{90} in soil is the sum of the concentrations of Sr^{90} mixed with calcium, S_m, and of Sr^{90} unmixed with calcium, S_u. The measured value of the strontium unit obtained from a sample of this soil can then be represented by

$$\text{measured S.U.} = \frac{S_m + S_u}{\text{Ca}} \qquad (3)$$

If we say that only the strontium mixed with calcium is applicable, then the strontium unit which should have been measured is

$$\text{applicable S.U.} = \frac{S_m}{\text{Ca}} \tag{4}$$

The applicable fraction of a given Sr^{90}/Ca ratio measured in soil is then

$$\frac{\text{applicable S.U.}}{\text{measured S.U.}} = \frac{S_m}{S_m + S_u} \tag{5}$$

Thus if all the Sr^{90} in soil is mixed with calcium, $S_u = 0$, the applicable fraction is unity, and all of the measured Sr^{90}/Ca ratio is significant. If none of the Sr^{90} is mixed with the soil calcium, $S_m = 0$, and none of the measured ratio is significant. With time, Sr^{90} freshly fallen on soil will mix with soil calcium, and S_m will increase as S_u decreases.

By means of simplifying assumptions, interesting values for the applicable fraction of ratios measured in soil may be derived: let the rate at which Sr^{90} atoms are added to soil be constant, and let the rate at which Sr^{90} atoms mix with calcium atoms in the soil be proportional to the number of Sr^{90} atoms in the soil which are not yet mixed with calcium. Specifically, let

S_u be the total unmixed Sr^{90} in soil at any time t;
S_m be the total mixed Sr^{90} in soil at any time;
k be the coefficient of mixing;
q be the rate at which unmixed Sr^{90} is added to soil;
λ be the physical decay constant for Sr^{90}.

The rate at which Sr^{90} appears *unmixed* in soil is expressed in equation (6), and the rate at which Sr^{90} appears *mixed* is expressed in equation (7):

$$\frac{dS_u}{dt} = q - kS_u - \lambda S_u \tag{6}$$

$$\frac{dS_m}{dt} = kS_u - \lambda S_m \tag{7}$$

As the conditions, we assume that S_u and $S_m = 0$ at zero time.

By application of standard methods for solving simultaneous equations, the following solution to the system is obtained:

$$S_u = \frac{q}{k + \lambda}\left(1 - e^{-(k+\lambda)t}\right) \tag{8}$$

$$S_m = \frac{q}{\lambda}\left(1 - e^{-\lambda t}\right) - \frac{q}{k + \lambda}\left(1 - e^{-(k+\lambda)t}\right) \tag{9}$$

The fraction of total Sr^{90} in the soil that is mixed with calcium is found by combining equations (8) and (9):

$$\frac{S_m}{S_m + S_u} = 1 - \frac{\lambda}{k + \lambda}\left(\frac{1 - e^{-(k+\lambda)t}}{1 - e^{-\lambda t}}\right) \tag{10}$$

By substituting for k (equal to $0.69/T_{1/2}$), the fraction may be evaluated. This was done for several "half-times for mixing" of Sr^{90} with calcium. Values for the fraction appear in Table 1.

Table 1. *Applicable fractions of* Sr^{90}/Ca *ratios measured in soil*

Years since fallout starts	Half-lives in years for mixing			
	1	12.5	25	50
1	0.3	0.05	0.04	0.001
2	0.5	0.07	0.06	0.03
5	0.7	0.1	0.08	0.04
10	0.8	0.2	0.1	0.07
25	0.9	0.4	0.3	0.2
50	0.9	0.6	0.4	0.2
100	1	0.6	0.5	0.3
200	1	0.7	0.5	0.3

It is interesting that if the mixing half-time is 50 years, only 0.3 of the measured ratio is applicable. This does not necessarily mean that the true value of any one ratio is three-tenths of the measured value of the ratio, for it is possible that the sample of soil was taken under such conditions as to yield the correct value. Instead, it means that if the mixing half-time is 50 years, then the ratio—even 100 years or more after fallout starts—is subject to a 300 per cent error. If, on the other hand, mixing occurs

rapidly—with a half-time of 1 year—the Sr^{90}/Ca ratios being measured in soil 10 years after fallout starts, for example, are subject to only a 20 per cent error.

In actual soil measurements, attempts are usually made to find "available" Sr^{90} and calcium by using an extraction procedure.[13] Although such a procedure tends to correct for different solubilities due to the different chemical forms in which the two elements may be present, it does not correct for differences in spatial distribution of the two elements in the soil. The roots of a growing plant do not necessarily absorb the same atoms that extraction makes available. Nevertheless, if the unmixed Sr^{90} is not extractable, extraction procedures will increase the applicability of the measured ratio.

In 1954, Hungate *et al.* initiated a series of field tests to evaluate some of the factors concerned in Sr^{90} availability to plants. As expected, movement down into the soil was a slow process and increased the uptake of Sr^{90} by the plants. Somewhat unexpected was the observation that Sr^{90} added to this calcareous soil 3 years previously was, by Neubauer tests in the laboratory, fully as available to plants as was Sr^{85} added just prior to cropping.[14] From this it appears that the time for Sr^{90} to achieve its ultimate equilibrium with calcium, in the sense of plant availability, is either very short or so long that three years is insignificant. A reduced availability of Sr^{90} was predicted from results of Martin *et al.*[15], who observed a decrease in labile Sr^{89} in soil with time, as measured by ammonium acetate extraction. It therefore appears that the mixing time is long and that the measurement of Sr^{90}/Ca ratios in soil to assess fallout hazards will lead to erroneous results.

If, instead of the Sr^{90}/Ca ratio, the Sr^{90}/Sr ratio is used (where Sr is the naturally occurring non-radioactive strontium), soil measurements would still be subject to error due to lack of mixing. It would still take time for Sr^{90} atoms to distribute themselves in soil to positions adjacent to the strontium atoms already present and time for chemical conversions to form compounds similar to those of the naturally occurring strontium.

Effect of environment

Most of the research which has been applied to understanding element-pair relationships involves testing the ability of the Sr^{90}/Ca ratio to qualify according to the third condition. Unlike cesium and potassium, strontium and calcium are only one period apart in the periodic table. It is reasonable to expect that strontium and calcium will behave more alike than cesium and potassium. The more alike a pair of elements are, the more probable it is that the ratios of the elements in two links of a biological chain will bear a constant relationship to one another. If deviations are found for the strontium–calcium pair, it is likely that greater deviations will be found for the cesium–potassium pair. The relationship expressed in equation (2) seems to imply that the *DF* is a proportionality constant, and is sometimes used as such.[17]

The results that led us to questioning the constancy of the *DF* were obtained in 1954 from plants. Rediske, Cline and Selders found no decreased uptake of Sr^{90} into plants when non-radioactive strontium was added to Sr^{90} in the root environment.[18] In 1956, Cline and Hungate got similar results when calcium atoms were used to dilute Sr^{90} atoms in soil. Again the uptake of Sr^{90} was not depressed by increasing soil calcium. It was about at this time that public interest in the hazards of fallout became widespread, and it was popularly asserted that the hazards of Sr^{90} was inversely proportional to the concentration of calcium in the environment. This was quite contrary to our results, at least in plants, and the difference between what was read in the papers and what was observed in the laboratory invited further investigation.

Before long, Hungate *et al.*[14], found that calcium could, indeed, depress the uptake of Sr^{90} into plants. However, this occurred only in acidic soils low in calcium. But even here a linear inverse relationship in depression of Sr^{90} uptake with increase of calcium was not noted. Fredriksson *et al.*[19], observed similar effects and pointed out that the uptake of Sr^{90} into plants is not inversely proportional to soil calcium.

The work with plants suggested that similar implications might result from work with

animals. In 1958, two feeding experiments were performed by PALMER *et al.* with mature rats. The first one involved only a 24-day feeding regimen[20] and will not be described here since its results are similar to those obtained in a second and more complete experiment.[21]

Four groups of mature, female rats were maintained for 30 days on diets containing 0.5 per cent phosphorus and four levels of calcium which was added as calcium lactate. After the conditioning period, Sr^{90}, Ca^{45} and unabsorbable Co_3O_4 tagged with Co^{60} were added to the diets. Four animals from each group were killed after 3, 6, 13, 24, 50 and 100 days on the tagged diets and their bones analyzed for Sr^{90} and Ca^{45}. Food consumption was estimated from measurements of Co^{60} in the total feces and gastrointestinal tract contents from the animals in each experimental group.

Had increases in dietary calcium caused proportionate decreases in Sr^{90} deposition, the results should have corresponded to the following equation, where the fraction of the dietary strontium that is deposited in bone is inversely related to the concentration of calcium in the diet:

$$\frac{Sr^{90}_{bone}}{Sr^{90}_{diet}} \simeq \frac{1}{Ca_{diet}} \qquad (11)$$

or

$$\frac{Sr^{90}_{diet}}{Sr^{90}_{bone}} = K_1 Ca_{diet} \qquad (12)$$

If equation (12) is to be obeyed, a plot of the reciprocal of the fraction of Sr^{90} deposited in the bone from the diet vs. the concentration of calcium in the diet should result in a straight line. Fig. 1 is such a plot based on the experimental results after 100 days of feeding. The reciprocal of the Ca^{45} deposition corresponds to nearly a straight line, but that for Sr^{90} deviates markedly.

The deviation observed from experiments performed with rats resembles that observed in plant experiments. Its implications with regard to the use of the strontium unit can be seen by expressing the results mathematically in the following two equations, where subscripts d and b refer to diet and bone. They state that the deposition of Ca^{45} on bone is inversely proportional to dietary calcium, but that the deposition of Sr^{90} is not. Instead, it is inversely proportional to some unspecified function of dietary calcium.

$$\frac{Ca^{45}_b}{Ca^{45}_d} = \frac{K_2}{Ca_d} \qquad (13)$$

$$\frac{Sr^{90}_b}{Sr^{90}_d} = \frac{K_1}{f(Ca)_d} \qquad (14)$$

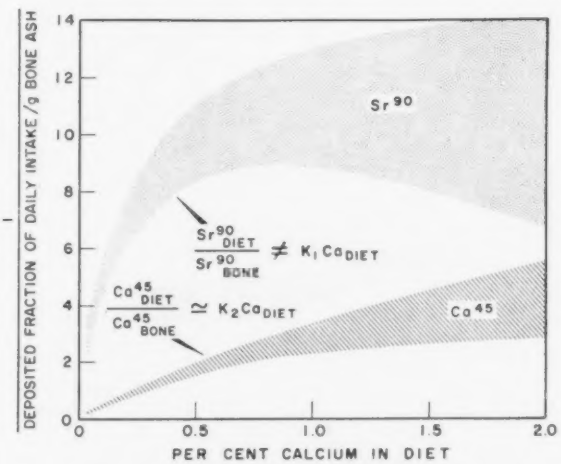


FIG. 1. Effect of dietary calcium on skeletal deposition of Sr^{90} in rats (width of lines proportional to standard deviations).

On dividing equation (14) by equation (13),

$$\frac{Sr^{90}_b/Ca^{45}_b}{Sr^{90}_d/Ca^{45}_d} = K\frac{f(Ca)d}{Ca_d} \qquad (15)$$

The result can be rearranged to

$$\left(\frac{Sr^{90}}{Ca^{45}}\right)_b = K\left[\frac{f(Ca)}{Ca}\right]_d\left(\frac{Sr^{90}}{Ca^{45}}\right)_d \qquad (16)$$

Equation (16) resembles equation (2), the definition of the *DF*. In this case, from equations (2) and (16),

$$DF = K\left[\frac{f(Ca)}{Ca}\right]_d \qquad (17)$$

This states that the *DF* is not a constant, but varies with dietary calcium. It resulted from the lack of linearity in the relationship between strontium deposition and the inverse of the dietary calcium concentration. In Fig. 2 are graphed *DF*s derived from the experiment in which rats were fed labeled strontium and calcium at various dietary levels of calcium.

Since our noting the variable nature of the *DF* contrasts with observations of others (for example, COMAR *et al.*[6]), who noted that the coefficient is relatively constant, it should be pointed out that an important difference between the experiments exists. The work that indicated a lack of variability with calcium utilized diets in which both dietary calcium and phosphate concentrations were varied to maintain a constant calcium to phosphorus ratio. In our work only the calcium was varied, with the phosphorus concentration maintained constant.

To find if the *DF* would change with dietary calcium in young rats, another experiment was performed by PALMER *et al.*[21]. Female rats, 22 days old, were conditioned for 4 days to

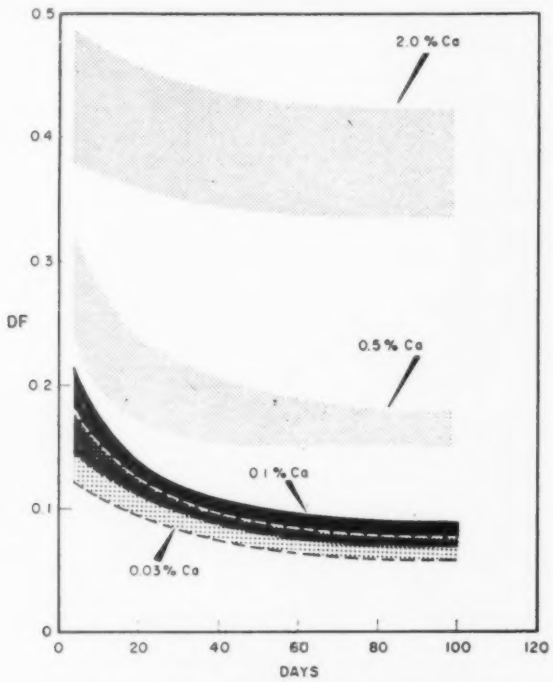


FIG. 2. Effect of dietary calcium on the *DF* (bone–diet) in mature animals at various times after feedings began (width of lines proportional to standard deviations).

diets like those used for the mature rats, except that only three levels of calcium were used—0.1, 0.5 and 2.0 per cent. Strontium-90 and Ca^{45} were then added to the diets and the feeding continued, with groups of six animals killed at intervals to 200 days on the labeled diets. Figure 3 was derived from the results.* It indicates that the *DF* (bone–diet) of these rats varied over a twofold range as a result of dietary calcium concentration and over nearly the same range as a result of the passage of time.

Whether the experimentally noted variability in the *DF* with calcium has significance in practical hazard assessment depends on the variation in dietary calcium among segments of population. Unfortunately, no data on variations are available. Data are available for the average diets, however. In a recent comprehensive study, HARLEY and RIVERA[22] estimated average intakes of various foods, calcium, and Sr^{90} by humans in the United States. The per cent of calcium in diets of infants during their first year of life averaged 0.18, in diets of adults, 0.06, and in diets of expectant mothers, from 0.03 to 0.07. These percentages, in combination with Figs. 2 and 3, suggest there is little reason to question the practical applicability of a constant number for the discrimination factor when used for average populations, at least so far as dietary calcium is concerned. Whether deviants from the average among the population of the United States and whether greater variations in dietary calcium among international populations would decrease the validity of the use of

* Due to undetermined causes, rats on the 0.1 per cent dietary calcium suffered high mortality, and too few were left near the close of the experiment to provide significant results at 200 days.

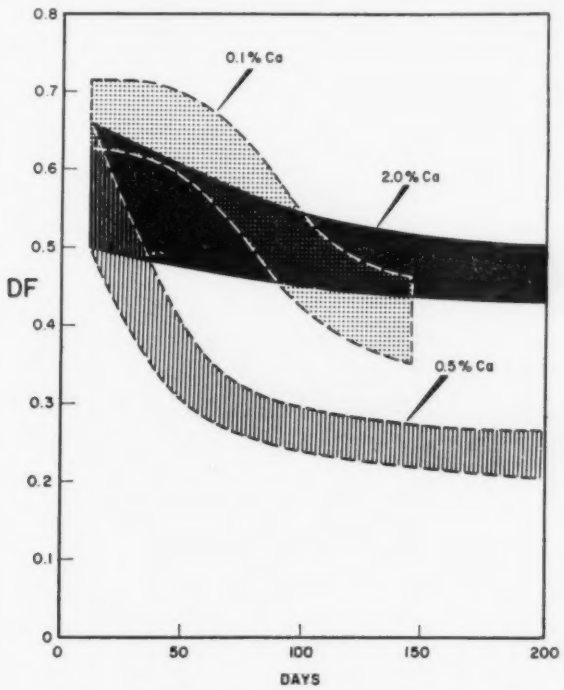


FIG. 3. Effect of dietary calcium on the *DF* (bone–diet) in growing animals at various times after feedings began (width of lines proportional to standard deviations).

a constant value for the discrimination factor is still unknown.

In a series of experiments to find if *DF*s in a plant would change, UHLER[23] grew red kidney bean plants in nutrient cultures containing Sr^{85}, Ca^{45} and several concentrations of calcium. He found the ratio in the leaves was the same as that in the substrate at all calcium concentrations tested. In the roots and stems, however, there was discrimination, as indicated in Fig. 4, where the *DF*s are shown as changing with change in the substrate calcium.

What applies to the strontium–calcium pair of elements should also apply to other pairs. It would seem that the use of the Cs^{137}/K ratio would be subject to more error than the use of the Sr/Ca ratio because cesium and potassium are more than one period apart in the periodic table. To test this possibility, CLINE[24] grew beans in nutrient solutions containing Cs^{137} and several concentrations of potassium. Although EPSTEIN and HAGEN[25] showed that potassium and cesium compete for deposition on excised barley roots, this competition was not apparent when the whole plant was tested. Instead, increasing concentrations of potassium had only a small effect on uptake of cesium. The relative independence of cesium and potassium thereby causes greater variations in the *DF* (plant–substrate) than were noted for strontium and calcium. The limitations to the use of the Cs^{137}/K ratio apparently are more stringent than those of the Sr^{90}/Ca ratio.

The contribution of dietary variables other than one member of the pair of elements toward making the *DF* a variable was sought in another animal experiment performed by PALMER *et al.*[26]. Six groups of mature rats were conditioned for 5 days to six diets which differed in calcium, phosphate, carbonate and lactate

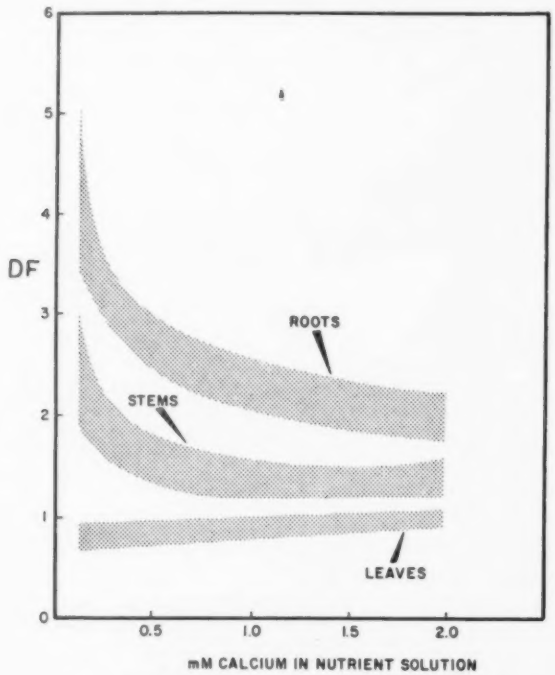


FIG. 4. Effect of substrate calcium on the *DF*'s (root-substrate, stem-substrate, and leaf-substrate) of bean plants (width of lines proportional to standard deviations.)

contents. Strontium-89 and Ca^{45} were added to the diets; 3 days later the rats were killed and their femurs analyzed for Sr^{89} and Ca^{45}. Table 2 shows that the *DF* changed particularly with change in carbonate.

It would be helpful to know why, in terms of fundamental processes, the *DF* varies with calcium concentration, since it is usually expected that such variation should not occur.

Table 2. Effect of some anions on the DF (bone–diet)

Per cent of dietary constituent tested			Calculated *DF*
Ca	P	CO_3	
0.1*	0.12	†	0.22
0.1	0.12	0.13	0.25
0.1	0.12	3.1	0.34
2.0*	0.12	†	0.60
2.0	0.12	3	0.46
2.0	2.4	3	0.41

* Calcium added as the lactate.
† Basal diet carbonate only.

The following explanation is offered more as an interim hypothesis to provoke other hypothesis and experimentation than to provide definitive explanations of the actual mechanisms involved.

It is proposed that the changing observed ratio is the consequence of transporting or depositing the pair of elements via two or more processes that operate in parallel, with the efficiency of at least one of them decreasing as the amount of one of the elements transported or deposited increases. For example, if the change with calcium of the *DF* in animals occurs as strontium and calcium are absorbed through the intestinal walls, then two or more processes transport the pair of elements through the gut wall and into the blood. The processes may consist of two or more substances that complex strontium and calcium and carry them into the blood stream. Similarly, if the *DF* changes because the kidney excretes different ratios of the two elements with change in calcium through-put, two or more parallel processes transport the elements from the blood to the urine. If the *DF* changes because the relative amounts of strontium and calcium that deposit on the bone change with calcium, then it can be explained by means of two or more deposition processes, such as those due to different kinds of bone crystals. In all cases, the theory requires that the effectiveness of at least one of them be decreased by increases in the quantity of calcium they transport or deposit.

How this model can account for a variable *DF* is seen by setting up hypothetical processes that can discriminate between strontium and calcium and then by defining the *DF* in terms of parameters used to describe the processes. Let it be assumed that discrimination occurs as strontium and calcium are absorbed from the lumen of the intestine, through the intestinal wall, and into the blood. This is schematically represented in Fig. 5, where an "active transport" process carries the strontium and calcium through the intestinal wall. The carrier substance *P* combines with the two elements to form products that can move from left to right in the diagrammatic representation of the intestinal wall. On the right-hand side the carrier is freed from strontium and calcium which enter the blood. Molecules of the carrier then return to the left where they once again combine with strontium and calcium to repeat the process.

If we say the Sr/Ca ratio in the lumen is the same as in the diet, the blood–diet discrimination factor for this case can be defined as

$$\frac{(\mathrm{Sr/Ca})_{\mathrm{blood}}}{(\mathrm{Sr/Ca})_{\mathrm{lumen}}} = DF \qquad (18)$$

The problem is then to see how increased calcium through-put, from lumen to blood in the model, can change the *DF*.

The quantity of strontium and calcium which can appear in the blood at any time depends on the efficiency with which the carrier *P* can combine with the elements—it is assumed that the process which limits transfer of strontium and calcium from the lumen to the blood is the rate at which the elements can be carried through the wall, and this depends on the concentrations of complexes formed by strontium and calcium combining with the carrier. It follows that the

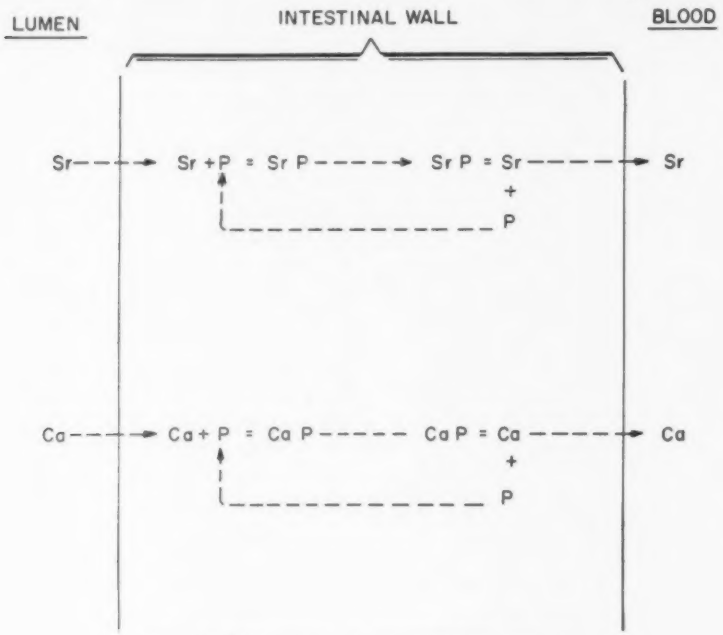


FIG. 5. Schematic of hypothetical carrier transport of strontium and calcium from the lumen of the intestine through the intestinal wall into the blood.

blood strontium and calcium will be proportional to the concentrations of SrP and CaP (see Fig. 5). As a result,

$$(\mathrm{Sr}/\mathrm{Ca})_{\mathrm{blood}} \simeq (\mathrm{Sr}P/\mathrm{Ca}P)_{\mathrm{wall}},$$

and, from equation (18),

$$DF = \frac{(\mathrm{Sr}P/\mathrm{Ca}P)_{\mathrm{wall}}}{(\mathrm{Sr}/\mathrm{Ca})_{\mathrm{lumen}}} \qquad (19)$$

To determine the DF in terms of measurable concentrations, the mass-action law can be used. The two reactions of strontium and calcium with the carrier are represented by the following equations:

$$\begin{array}{cccc} \mathrm{Sr} + P = \mathrm{Sr}P & & \mathrm{Ca} + P = \mathrm{Ca}P \\ s \quad p - v \quad u & & c \quad p - v \quad v \end{array}$$

$$K_1 = \frac{u}{s(p - v)} \qquad K_2 = \frac{v}{c(p - v)} \qquad (20)$$

Underneath the symbols in the two equations are written steady state concentrations of elements, the carrier, and the products. For every equivalent of product, CaP, formed there will be one less equivalent of the carrier P available. Thus if v equivalents of CaP are formed and if p is the *original* (or *total*) number of equivalents of carrier, the number of equivalents of P *available* for complexing with Sr and Ca is $p - v$. (The steady-state concentration of P need not be $p - u - v$ because u, the number of equivalents of SrP, is insignificant in comparison with the number of equivalents of CaP.)

By substituting in equation (19) the steady-state concentrations of products and reactants given above,

$$DF = \frac{u/v}{s/c} \qquad (21)$$

But these concentrations are interrelated according to the expressions for the steady-state

constants, K_1 and K_2. Upon substituting for u, v, s, and c from the two expressions in (20),

$$DF = \frac{K_1}{K_2} \tag{22}$$

There is no variable in this expression; K_1 and K_2 are constants. In this model, if there is only one process (one carrier, for example) for transferring strontium and calcium into the blood, the *DF* cannot change with change in calcium. Another process, operating in parallel with the one given, will give the required result. The second process could consist of another carrier or be simple diffusion. Either way, the discrimination factor becomes a function of the calcium concentration in the lumen.

If it be assumed that another substance Q is present to combine with strontium and calcium forming complexes that can be transported through the intestinal wall, acting similar to the carrier P, a new expression for the *DF* can be derived by again considering mass-action formulations. The reactions involved, steady-state concentrations, and definitions of steady-state constants follow:

$$\underset{s\quad q\quad\ x}{\mathrm{Sr} + Q = \mathrm{Sr}Q} \qquad \underset{c\quad q\quad\ y}{\mathrm{Ca} + Q = \mathrm{Ca}Q}$$

$$K_3 = \frac{x}{sq} \qquad K_4 = \frac{y}{cq} \tag{23}$$

In this second process the capacity of the carrier Q is not assumed approached; its steady-state concentration is given as q, rather than as $q - y$, which would be equivalent to the case of the first carrier, P. This was done to show that although two (or more) parallel processes for transporting strontium and calcium can account for a variable discrimination factor, it is not required that both of them approach saturation.

The *DF* must now be evaluated in terms of the *sum* of the concentrations of strontium and calcium that are complexed with the two carriers. This is

$$DF = \frac{(u + x)/(v + y)}{s/c} \tag{24}$$

The concentrations on the right side of the equation are interrelated according to the expressions for the four steady-state constants, K_1, K_2, K_3 and K_4. Upon substituting into equation (24) values for u, x, v and y from equations in (20) and (23),

$$DF = \frac{K_1 p + K_3 q(1 + K_2 c)}{K_2 p + K_4 q(1 + K_2 c)} \tag{25}$$

Equation (25) expresses the *DF* in terms of constants and the concentrations of calcium in the diet of the model, and suggests that it is reasonable to expect the *DF* to vary with concentration of dietary calcium. The equation is of the form

$$y = \frac{A + Bx}{C + Dx} \tag{26}$$

where y is the discrimination factor, x is the concentration of calcium in the lumen of the intestine, and A, B, C and D are constants. It is the equation of a hyperbola which consists of two conjugate pairs of curves. On rectangular co-ordinates, one pair occurs in the first and third quadrants, and the other in the second and fourth quadrants. Of these four only two have physical meaning—those which occur in the first and second quadrants—because they correspond to positive values of x. The other two correspond to negative values of x, or negative concentrations of calcium, which has no physical meaning, and will be ignored.

Figure 6 is a plot of equation (26). The two curves that approach a common asymptote, $y = B/D$, result from the relationship of the constants to each other. If A/C is greater than B/D, the upper curve results. If it is less than B/D, the lower curve results. In Fig. 6, the two values for A/C corresponding to each curve are shown as subscripts.

The hypothesis of two parallel processes operating to transfer strontium and calcium predicts that the discrimination factor should either increase or decrease with increase in calcium concentration and that the change should be greatest at low calcium concentrations, tending, at high calcium concentrations, to approach a constant number. If the hypothesis is correct, and if all plants and animals lived in a calcium-rich environment, the yet-to-be-determined experimental asymptote value of the *DF* could be used for predicting Sr^{90}

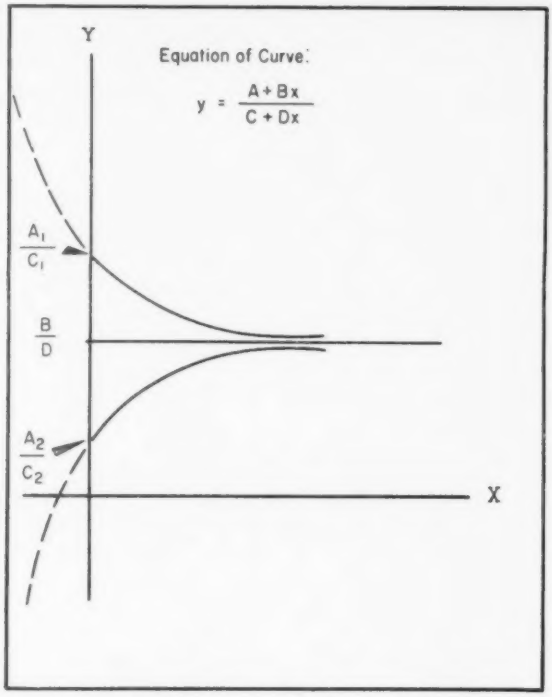


FIG. 6. A plot of the theoretical equation for the discrimination factor (y) changing as a function of the calcium concentration (x).

contamination with accuracy, at least so far as environmental calcium is concerned. But only presumptive evidence suggests the hypothesis is correct. The upper curve in Fig. 6 resembles part of Fig. 4, where the *DF* in roots and stems of bean plants decreased with increasing substrate calcium. When the *DF*s obtained at 100 days from the prolonged feeding experiment with rats previously described[19] and illustrated in Fig. 2 were plotted vs. dietary calcium, Fig. 7 resulted. This suggests the lower theoretical curve in Fig. 6.

There is not enough information to tell whether similarities of theoretical and experimental results represent happy coincidence or support of theory. More work designed to investigate the mechanisms is needed. First, we need to find which of those organs or tissues in the animal body that can discriminate between strontium and calcium will change their degree of discrimination with change in concentration or through-put of calcium. Once found, it will be interesting to see if there are, indeed, two or more processes that carry or deposit strontium and calcium in such tissues. If there are, it should be possible to determine if the two or more processes are due to different kinds of complexing agents for carrying the pair of elements or whether there is but one kind of carrier and the parallel process is but simple diffusion.

Since increments of dietary calcium do not cause proportionate decrements of Sr^{90} on bone, it is pertinent to ask what happens to Sr^{90} deposition with increase in non-radioactive strontium. The difficulty with answering the question experimentally is that the quantity of non-radioactive strontium which is added to the diet to get an effect must be significant in comparison with the quantity of calcium

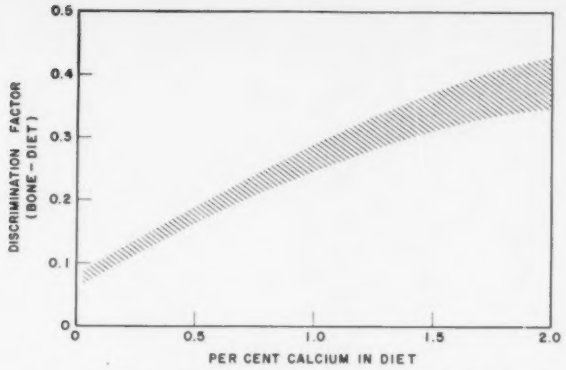


FIG. 7. The change in the discrimination factor with dietary calcium after 100 days of feeding strontium and calcium to rats.

already present in the diet. Or conversely, dietary calcium must be kept to practically zero levels in order to make small increments of non-radioactive strontium effective in decreasing Sr^{90} deposition. It is the sum of the quantities of calcium-like elements that count, and strontium is like calcium in this regard.

The importance of the sum of calcium-like elements was shown by UHLER[23] in an experiment with plants. Cultures of beans were grown in nutrient solution containing Sr^{90} and Ca^{45} and different ratios of non-radioactive strontium to non-radioactive calcium, with the sum of the molar concentrations of the two elements remaining the same in all cultures. After the growth period, uptake of Sr^{90} and Ca^{45} was measured. Except for slight changes at high strontium concentrations due to toxicity, the *DF* was unaffected by changing the ratio. This indicates that it is the sum of the number of moles of strontium plus calcium that is relevant in changing uptake of Sr^{90} and Ca^{45}.

A similar experiment in animals may be impossible, due to the toxicity of strontium when present in the diet in quantities significant in comparison with calcium. If it could be performed, the results should be similar to those observed in plants. But in animals fed calcium within the normal dietary range, any practical quantity of non-radioactive strontium added to the diet should be without effect in changing Sr^{90} deposition because the total alkaline earth concentration in the diet is not significantly changed.

Effect of time

That the Sr^{90}/Ca ratio may not meet the fourth condition is suggested from Figs. 1 and 3 where the *DF* changes with time. If it does change, it means that the rates of deposition of strontium and calcium on to, or the rates of release from, bone were changing with time. To find whether the rates of release from bone of the two elements differed, THOMPSON and PALMER measured their retention on the bone with time.[27] Groups of mature female rats, previously conditioned for 28 days on diets containing 0.03, 0.1, 0.5 and 2.0 per cent calcium, were intraperitoneally injected with Sr^{90} and Ca^{45} and maintained on the same diets for 200 days. During this period animals from each dietary group were killed at various times after the single injection of radioelements and their femurs analyzed for Sr^{90} and Ca^{45}. The results appear in Fig. 8. Several interesting relationships are evident.

First, dietary calcium changed the slope of the turnover curves for deposited Ca^{45}, but was without apparent effect on Sr^{90}. It is of incidental interest that this point tends to deny the possibility of speeding the removal of strontium from the skeleton by increasing dietary calcium. Second, the turnover from bone of both strontium and calcium can be described

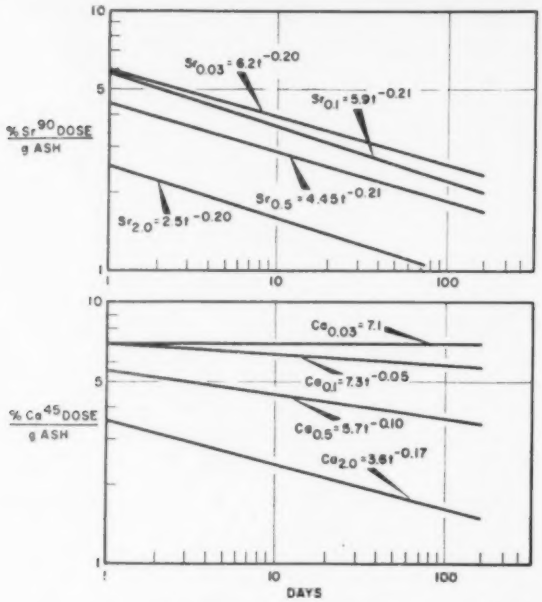


FIG. 8. Logarithmic plot of amounts of Sr^{90} and Ca^{45} remaining in bone vs. time after single IP injections of the radioelements (subscripts following symbols of elements refer to dietary calcium levels).

by power functions of time since the data, plotted on logarithm paper, fit straight lines. Thus the concentrations in the bone of the two radioelements at any time t during the period of observation (200 days) can be represented by

$$\mathrm{Sr} = At^{-m} \quad \text{and} \quad \mathrm{Ca} = Bt^{-n} \tag{26}$$

where A and B are the concentrations of the two radioelements when $t = 1$ day and m and n are slopes of the curves.

The two equations in (24), descriptive of the concentrations in bone of strontium and calcium at time t after a single administration of the two elements, can be integrated to yield equations that describe the concentrations of the two elements in bone with time after continuous administration begins. These are

$$\mathrm{Sr} = \frac{At^{1-m}}{1-m} \quad \text{and} \quad \mathrm{Ca} = \frac{Bt^{1-n}}{1-n} \tag{27}$$

(The values of m and n are obviously restricted to being greater than zero, but less than 1.)

On dividing the first equation by the second,

$$\frac{\mathrm{Sr/Ca}}{A/B} = \frac{1-n}{1-m}\, t^{n-m} \tag{28}$$

By definition [equation (2)], the discrimination factor is

$$DF = \frac{(\mathrm{Sr/Ca})_b}{(\mathrm{Sr/Ca})_d} \tag{29}$$

On substituting for $(\mathrm{Sr/Ca})_b$ from equation (26),

$$DF = \frac{A/B}{(\mathrm{Sr/Ca})_d} \cdot \frac{1-n}{1-m}\, t^{n-m} \tag{30}$$

In going from equations (26) to (30), we show, in effect, that if the retentions of strontium and calcium can be described by different power functions of time, the DF changes with time. THOMPSON and PALMER substituted into equation (30) values for coefficients found from the single dose retention experiment and got a plot of DF curves that coincided with the

experimental *DF* curves obtained when rats were allowed to continuously feed on diets containing the two radionuclides.[27]

This implies that the Sr^{90}/Ca ratio in the skeleton of individuals should change with time even though his diet contains an unchanging Sr^{90}/Ca ratio. Consequently, it implies also that the natural strontium to calcium ratio should change with time in individuals on diets containing unvarying ratios of these elements. BRYANT *et al.*[28], after analyzing human bone for both radioactive and natural strontium, reported that the natural strontium to calcium ratio increased with age. Although this might be due to changes in diet with age, in view of our experimental observations, it may be due to continuing change with age in the net skeletal deposition of strontium and calcium. Resolution of this point will depend on showing whether the net skeletal deposition of strontium and calcium ceases to change with time sufficiently early in life.

CONCLUSIONS

In using measurements of ratios of similar elements in the biosphere to predict future ratios, potential sources of error result from the way biological systems apparently can change their discriminatory powers. The principal causes for such changes are unequal availability of the two elements, different environments, and the passage of time. Whether variability of discrimination is significant in practical radiation hazard evaluation is unknown, but there is sufficient evidence to indicate that caution should be exercised in using ratios.

It may be simpler (and less reassuring) to express Sr^{90} contamination in terms of conventional concentration units. Until more information becomes available to support one or the other method, it is recommended that Sr^{90} contamination levels, both measured and predicted, be given in terms of conventional concentration units as well as in terms of the ratio with calcium.

Even on the basis of fragmentary evidence, the vagaries of the Cs^{137}/K unit contra-indicate its continued use. All the weaknesses of the Sr^{90}/Ca ratio seem to be magnified in the Cs^{137}/K ratio. There is nothing inherently wrong with measuring Cs^{137} contamination in terms of the ratio, but there appears little to support its use in predicting contamination levels.

REFERENCES

1. R. S. CALDECOTT and L. A. SNYDER, *A Symposium on Radioisotopes in the Biosphere* pp. 255–268. University of Minnesota (1960).
2. J. L. KULP, A. R. SCHUBERT and E. J. HODGES, *Science* **132,** 448 (1960).
3. E. C. ANDERSON, *Science* **128,** 882 (1958).
4. *Worldwide Effects of Atomic Weapons*, U.S. Atomic Energy Commission Report R-251, p. 73 (1953).
5. C. L. COMAR, I. B. WHITNEY and F. W. LENGEMANN, *Proc. Soc. Exp. Biol., N.Y.* **88,** 232 (1955).
6. C. L. COMAR, R. H. WASSERMAN and M. M. NOLD, *Proc. Soc. Exp. Biol. Med., N.Y.* **92,** 859 (1956).
7. C. L. COMAR, R. H. WASSERMAN, S. ULLBERG and G. A. ANDREWS, *Proc. Soc. Exp. Biol., N.Y.* **95,** 386 (1957).
8. C. L. COMAR, R. SCOTT RUSSELL and R. H. WASSERMAN, *Science* **126,** 485 (1957).
9. G. V. ALEXANDER, R. E. NUSBAUM and N. S. MACDONALD, *J. Biol. Chem.* **218,** 911 (1956).
10. R. G. MENZEL, *Soil Sci.* **77,** 419 (1954).
11. W. H. LANGHAM and E. C. ANDERSON, *Health Phys.* **2,** 30 (1959).
12. R. SCOTT RUSSELL, *Nature, Lond.* **182,** 834 (1958).
13. H. J. M. BOWEN and J. A. DYMOND, *J. Exp. Bot.* **7,** 264 (1956).
14. F. P. HUNGATE, R. L. UHLER and J. F. CLINE, *Radiostrontium uptake by plants.* In Hanford Biology Research Annual Report. Document HW-53500, pp. 7–17 (1958).
15. R. P. MARTIN, P. NEWBOULD and R. SCOTT RUSSELL, Discrimination between strontium and calcium in plants and soils. Lecture during the UNESCO International Conference, Paris, France, 9–20 September (1957).
16. R. H. WASSERMAN, C. L. COMAR and D. PAPADOPOULOU, *Science* **126,** 1180 (1957).
17. UN General Assembly, 1958, Report of the United Nations Scientific Committee on the effects of atomic radiation. Official Records: Thirteenth Session Supplement No. 17 (A/3838), 114 New York (1958).
18. J. H. REDISKE, J. F. CLINE and A. A. SELDERS, *The Absorption of Fission Products by Plants.* In Hanford Biology Annual Report for 1954, Document HW-35917, pp. 40–46 (1955).
19. L. FREDRIKSSON, B. ERIKSSON, B. RASMUSON, B. GAHNE, K. EDVARSON and K. LÖW, Studies

on soil-plant-animal interrelationships with respect to fission products. *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1955, Vol. 18, pp. 449–470. United Nations, New York (1956).

20. R. F. Palmer, R. C. Thompson and H. A. Kornberg, *Science* **127,** 1505 (1958).
21. R. F. Palmer, R. G. Thompson and H. A. Kornberg, *Strontium–Calcium Interrelationships in the Mature and Growing Rat.* In Hanford Biology Research Annual Report for 1958, Document HW-59500, pp. 20–28 (1959).
22. J. H. Harley and J. Rivera, *Summary of Available Data on the Strontium^{90} Content of Foods and of Total Diets in the United States.* USAEC Document HASL-90 (1960).
23. R. L. Uhler, *Influence of Calcium on Radiostrontium Uptake.* In Hanford Biology Research Annual Report for 1958, Document HW-59500, pp. 20–28 (1959).
24. J. F. Kline and F. P. Hungate *Plant Physiol.* **35,** 826 (1960).
25. E. Epstein and C. E. Hagen, *Plant Physiol.* **27,** 457 (1952).
26. R. F. Palmer, R. C. Thompson and H. A. Kornberg, *Science* **128,** 1505 (1958).
27. R. C. Thompson and R. F. Palmer, *Amer. J. Physiol.* **199,** 94 (1960).
28. F. J. Bryant, J. C. Cotterill, E. H. Henderson, G. S. Spicer and T. J. Webber, *Radioactive and Natural Strontium in Human Bone.* UKAEA Document AERE-R-2988 (1959).

Health Physics Pergamon Press 1961. Vol. 6, pp. 63–65. Printed in Northern Ireland

THE ANALYSIS OF PLUTONIUM-241 IN URINE

J. DONALD LUDWICK*
Hanford Laboratories Operation, General Electric Company, Richland, Washington

(*Received* 14 *March* 1960)

Abstract—A method has been developed for the quantitative determination of plutonium-241 in exposed personnel through urine analysis, and was designed for use, when necessary, in conjunction with the Hanford plutonium bio-assay technique of α-analysis. This method is based on the recovery of plutonium from electro-deposited disks and subsequent extraction of the plutonium from 1 N hydrochloric acid into a liquid scintillator containing dibutyl phosphate. Plutonium-241 β-emission is detected in a standard liquid scintillation spectrometer, and the yield for plutonium recovery using spiked samples of Pu^{241} was 85 ± 9 per cent. Samples containing as little as 2.2×10^{-6} μc of Pu^{241} may be reliably detected, which represents 3.5 per cent of the maximum permissible dose 90 days after personnel exposure.

INTRODUCTION

Processing of high-exposure uranium and plutonium fuels may result in potential exposure of chemical processing personnel to a number of heavy plutonium isotopes. All of these isotopes can be determined by α-counting techniques with the exception of the β-emitter Pu^{241}. Production of this isotope is about 10 per cent of the total plutonium, predicted after only the third cycle or approximately one year of operation of the Plutonium Recycle Test Reactor at Hanford. Plutonium-241 will then be present in amounts large enough to contribute significantly to the exposure received in internal contamination. The work herein described was carried out in order to provide an analytical procedure for monitoring individuals potentially exposed to the Pu^{241} isotope.

Utilization of excretion analysis for the quantitative determination of total body burden in humans involves the relationship between deposition and excretion level as a function of time. In many cases, it is customary to express isotope excretion as a simple exponential function of time, consequently arriving at a biological half-life. This presumes that the rate of elimination of an isotope from the body is controlled by a single body function. Often this assumption is not valid as in the case of tritium; here, the rate of excretion in the mouse appears to be controlled by three body functions resulting in half-times of 1.9, 12 and 90 days[1,2].

The rate of human urinary excretion of plutonium has been studied for a period of 5 years and found to be continually changing and best represented by a power function[3]. This is thought to be the result of several plutonium depositories in the body each eliminating plutonium at differing rates. The expression given for activity elimination per 24-hr day is

$$A = 0.002\ T^{-0.74} \quad \text{(initial dose)} \qquad (1)$$

where T = time in days since plutonium intake. Integration of this equation allows one to estimate the total plutonium excretion between two time limits, while solution of the expression for the 50 per cent excretion time indicates that 200 years are required for elimination of half the human body burden.

A monitoring program was needed to identify exposed personnel who may approach the maximum permissible dose over a period of time. The maximum permissible body burden for Pu^{241} has been set at 0.9 μc[4]. It was considered necessary that the sensitivity of the beta counting procedure be able to detect at least 5 per cent (corresponding to 3.2×10^{-6} μc/24-hr urine sample) of the maximum permissible body

* Work performed under Contract No. AT(45-1)-1350 for the United States Atomic Energy Commission.

burden 90 days after intake. This method must also be compatible with the already existing bio-assay procedure for the determination of plutonium by alpha-particle detection[5]. This latter procedure involves the chemical separation of plutonium from urine samples followed by electro-deposition of the plutonium in a known area on disks. Nuclear-track techniques are then utilized to detect as little as 0.05 dis/min of plutonium α-activity by using an exposure time of about 5 days. Subsequent to α-analysis the plutonium is probably in the form of PuO_2 in a highly insoluble film on a stainless steel disk.

EXPERIMENTAL

The maximum energy of the β-particles associated with Pu^{241} is 20 keV. Although there are numerous detection problems associated with this weak β-energy, the technique of liquid scintillation counting has been shown to be desirable for Pu^{241} detection[6].

Samples of urine were spiked with Pu^{241} whose β/α ratios were 27:1 and 1.6:1 (as the result of the alpha-emitting isotopes Pu^{239}, Pu^{240} and Pu^{242} present) and processed through the bio-assay procedure. Their α-activity was determined using zinc sulfide scintillation detectors, and the yield from the bio-assay procedure was found to be about 90 per cent. Then the stainless steel discs were placed in 5-ml beakers and treated with concentrated nitric acid containing a few drops of concentrated hydrofluoric acid. The clean disk was removed from the acid and the solution evaporated to near dryness under a heat lamp. Concentrated sulfuric acid was then added and the film dissolved under fuming action. This often required fuming times of 1 hr or more. When the film was completely dissolved and the acid volume small, the solution was diluted with 5 ml of 2 N nitric acid and transferred to a polystyrene test tube. The addition of 25 λ of lanthanum nitrate carrier solution (80 g/l.) and excess concentrated hydrofluoric acid resulted in fluoride precipitation. The plutonium was carried with the precipitate which was redissolved in dilute nitric acid and the process repeated. Iron is removed in this process so that it will not interfere with the oxidation state of the plutonium or discolor the liquid scintillator. The fluoride precipitate is again dissolved in 2 N nitric acid and transferred to a 5 ml beaker. The volume is reduced under a heat lamp and concentrated perchloric acid added. The sample was fumed to dryness to remove the fluoride and oxidize the plutonium to the +6 state. Other oxidation techniques were employed; however, the greatest extraction yield was obtained using perchloric acid. The plutonium was dissolved in 1.2 N hydrochloric acid and extracted with dibutyl phosphate (DBP). This latter step is very dependent upon the hydrochloric acid concentration. The extraction coefficient of the system DBP–HCl for plutonium has a maximum value at about 1 N hydrochloric acid. It is important to keep the concentration of hydrochloric acid above 0.5 N to prevent other plutonium reactions. Normally, the plutonium activity is removed from the beaker with small portions of 1.2 N hydrochloric acid and transferred to a 1-ml volumetric flask. To this, 10λ of DBP is added along with 100λ of a liquid scintillator. The scintillator solution found to dissolve DBP and be most efficient for β-detection was p-terphenyl (4.0 g/l.) and 1:4-bis-[2-(5-phenyloxazolyl)]-benzene (POPOP) (0.1 g/l.) in toluene. The two phases were shaken for about 3 min and subsequently centrifuged. The organic phase was withdrawn and placed in an 8-mm quartz vial. The extraction step was repeated three times and the organic phases combined in the vial, giving a final total volume of 0.4 ml of liquid scintillator solution. The quartz vial was sealed under vacuum to reduce dissolved gas quenching in the liquid scintillator. The samples were then placed in a Lucite light-pipe which is coupled to the multiplier phototubes of a liquid scintillation spectrometer, Tri-Carb, Model 314, Packard Instrument Company, LaGrange, Illinois. The samples were counted for about 1 day after allowing them to cool and stabilize in a dark room for about 1 hr.

RESULTS

Experimental results of the Pu^{241} β-analysis are shown in Table 1 along with the over-all yield from processing the disks into the liquid scintillator. The standard deviations of the individual measurements are also included. The low-activity samples were counted about 24–60 hr while higher activity samples were counted approximately 2 hr. For the former counting time, samples of Pu^{241} β-activity may be reliably detected down to at least 2.2×10^{-6} μc. This represents about 3.5 per cent of the maximum permissible dose 90 days after intake. These results were obtained without interference to the already existing bio-assay procedure, and the β-technique may be accomplished harmoniously in conjunction with the alpha analysis.

The body burden, B_m, at the time of exposure following a single occurrence may be calculated

Table 1. *The observed activity of samples before and after processing*

Sample (net)	α-Scintillation counting rate (dis/min)	Liquid scintillation counting (β^- counts/min)	(α dis/min)	(β^- dis/min)	Process yield (%)	β/α theoretical	Ratio calculated
1A	2.60	16.61 ± 0.29	2.05 ± 0.10	55.36 ± 0.96	79	27:1	27:1
2A	219.94	1383 ± 5	179.5 ± 1.7	4609 ± 16	82	27:1	26:1
303	4.28	2.08 ± 0.09	4.01 ± 0.06	6.49 ± 0.30	94	1.6:1	1.7:1
305	6.60	2.69 ± 0.24	4.99 ± 0.16	8.98 ± 0.80	76	1.6:1	1.8:1
306	3.10	1.22 ± 0.10	2.51 ± 0.80	4.06 ± 0.33	81	1.6:1	1.6:1
Background	0.00	0.00 ± 0.06	0.00 ± 0.05	0.00 ± 0.20	—	—	—
Control	3.34	0.63 ± 0.08	2.77 ± 0.06	2.11 ± 0.27	83	—	—
Control	0.00	0.44 ± 0.07	0.00 ± 0.05	1.48 ± 0.23	—	—	—

from the assay of a 24-hr urine specimen after time T using equation (1),

$$B_m = 500\, A\, T^{0.74} \tag{2}$$

More commonly, one is exposed to an unknown variable dose since the time of the last negative analysis. The body burden B_m, may be calculated in this case by assuming a single effective dose occurring at a time which can be determined from assay of two samples taken at a time X apart[3].

The activities in the first and second urine samples collected after time T and $T + X$, respectively, are

$$A_T = 0.002\, B_m'\, T^{-0.74} \tag{3}$$

$$A_{T+X} = 0.002\, B_m'\, (T + X)^{-0.74} \tag{4}$$

Solving for T,

$$\left(\frac{A_T}{A_{T+X}}\right) = \left(\frac{T}{T+X}\right)^{-0.74} \tag{5}$$

$$T = \frac{X}{(A_T/A_{T+X})^{1.35} - 1} \tag{6}$$

The body burden B_m' from equation (3) is then

$$B_m' = 500\, A_T \left[\frac{X}{(A_T/A_{T+X})^{1.35} - 1}\right]^{0.74} \tag{7}$$

The liquid scintillation spectrometer has sufficient resolution for separating alpha activity and the weak Pu^{241} β-activity. Where higher α-activity is present this method may be used for simultaneous α- and β-analysis; however, where very low α-activity is present (< 1.0 dis/min), the liquid scintillator has its limitations.

The gain of the instrument used was adjusted so that 30 per cent of the plutonium-241 β-spectrum was counted. Under these conditions the background counting rate for the β-region was 17 counts/min while the α-counting region had a background of 6 counts/min.

The experimental procedure described in this report does not add any α- or β-activity to the sample as evidenced by the background counting rate illustrated in Table 1, but it is apparent from control sample information that some beta activity is introduced in the chemicals used during the α bio-assay analysis.

In emergency cases of high personnel exposure or where α-analysis is unnecessary, this Pu^{241} β-analysis prodecure may be used without going through the original α bio-assay analysis. Immediate urine sampling may be instituted when Pu^{241} is suspected of being introduced into the body. The Pu^{241} may be counted after chemical separation and subsequent DBP extraction into the liquid scintillator. The β-activity will be considerably higher because of the short delay between exposure and sample collection. The sensitivity of the method is not affected by this change in procedure, however, as little as 0.1 per cent of the excreted activity can be detected one day after intake for personnel acquiring the maximum permissible dose.

REFERENCES

1. E. A. Pinson, LA-1467 (1952).
2. R. C. Thompson, *J. Biol. Chem.* **197,** 81 (1952).
3. W. H. Langham, *Amer. Industr. Hyg. Ass. Quart.* **18,** 305 (1956).
4. U.S. Department of Commerce, National Bureau of Standards Handbook 69 (1959).
5. L. C. Schwendiman and J. W. Healy, *Nucleonics* **16,** 78 (1958).
6. D. L. Horrocks and M. H. Studier, *Anal. Chem.* **30,** 1747 (1958).

Health Physics Pergamon Press 1961. Vol. 6, pp. 66–85. Printed in Northern Ireland

DISPERSION OF RADIOCONTAMINANTS IN AN ESTUARY*

F. L. PARKER†‡, G. D. SCHMIDT§, W. B. COTTRELL** and L. A. MANN**
Health Physics Division, Oak Ridge National Laboratory††,
Oak Ridge, Tennessee

(*Received* 1 *November* 1960)

Abstract—The dispersion of radioactive contaminants in an estuary is more complex than the dispersion in a flowing fresh-water stream due to tidal cycling in the estuary. Many approximate, analytical solutions are available for the hydraulic dispersion, but a model study is more accurate. The model study, however, ignores radioactive decay, and the absorption and adsorption of the nuclides by the sediments and the biota.

An evaluation of the maximum credible accident to the NS *Savannah* during start-up in the Delaware River shows that hazardous conditions will not exist. The evaluation is based on model studies by the U.S. Army Engineer Waterways Experiment Station and includes the nuclear decay and sorption of the nuclides. The maximum credible accident assumes 600 days operation at 69 MW and that 100 per cent of the fuel rods fail. It is also assumed that only a percentage of the isotopes will escape from the containment vessel, at a leakage rate of 0.2 per cent of the free volume of the containment vessel per day. Exposures from swimming and boating in the water and drinking the water from the river are evaluated. The maximum hazard occurs during the few days that the maximum concentration in the river water is about twice the maximum permissible concentration for continuous occupational exposure.

INTRODUCTION

In 1955 President Eisenhower proposed the construction of a nuclear-powered passenger and cargo ship, now called the NS *Savannah*, as a symbol of this country's interest in the peaceful uses of atomic energy. The ship was launched at the Camden, New Jersey, facility of the New York Shipbuilding Corporation on 21 July, 1959, and has been outfitted with a reactor which is scheduled to go critical the latter part of 1961. The ship is expected to tour the world as part of the U.S. Atoms for Peace program, and thus, must be as hazard-proof as possible.

In particular, this paper will deal with the problem of releases of activity to an estuary. An estuary is defined as a passage where the tide meets the river current; especially, an arm of the sea at the lower end of a river. Many of the world's major ports as well as the start-up site of the NS *Savannah* at Camden, New Jersey, are located on estuaries.

DISPERSION OF RADIOACTIVE MATERIAL

The fate of radioactive material introduced into a stream is affected by convection, diffusion, sorption, sedimentation and radioactive decay.

Convection

In a non-tidal stream, convection terms can be readily obtained from velocity profiles of the stream. In a tidal stream, however, the situation is far more complex, as there is alternately convection upstream and downstream. Measurements in the field are extremely difficult to make in the tidal sections of the river as is evidenced by the fact that the U.S. Geological Survey, the official stream-gaging organization, does not measure flow in the tidal portion of the

* Based upon ORNL-2867, *Environmental Analysis of NS Savannah Operation at Camden.*

† Member of Health Physics Division, ORNL.

‡ Present address: International Atomic Energy Agency, Vienna, Kaerntnerring, Austria.

§ Member of the U.S. Public Health Service.

** Member of the Reactor Projects Division, ORNL.

†† Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

stream, although it does measure the velocities. The usual method of determining flow in a tidal stream is to measure the flow in the non-tidal portions and then assume the same net inflow per unit of drainage area in the tidal portion as in the non-tidal portion. Another method is to simulate the stream by a model complete with tides and stream velocities and determine the flow from the model.

Diffusion

The contaminant is also dispersed in a stream by diffusion. The diffusion process includes both molecular and turbulent diffusion. Molecular diffusion is the result of the thermal motion of individual molecules and is extremely slow in liquids and gases, because the mean free path of molecules is so short. Turbulent diffusion may be much faster than molecular diffusion, since it is due to the mass transfer of molecules in eddies of various sizes throughout the entire volume. Molecular diffusion is essentially submicroscopic in movement, while turbulent diffusion is macroscopic.

The radioactive material is presumed to behave dynamically (diffuse) in the same way as the transporting medium—water. The radioactive substance is treated as radioactive water dispersed in water of no radioactivity. Many experiments support the validity of this assumption.[1]

The differential equation for diffusion is relatively simple if the diffusion coefficient remains constant throughout the region of flow:

$$\frac{\partial c}{\partial t} = \alpha^2 \frac{\partial^2 c}{\partial x^2} \qquad (1)$$

where c = concentration;
α^2 = diffusion coefficient;
x = length;
t = time.

The standard formulation of the diffusion law is commonly known as Fick's law. Eddy diffusion has been found to vary from 1000 to 1,000,000 times greater than molecular diffusion in water.[2] Therefore, molecular diffusion can be ignored.

In the ocean, turbulent diffusion in large areas may be taking place in numerous ways within the same area. Large- and small-scale eddies within them induce various rates of mixing. The complete randomness of the process makes it a very difficult phenomena to formulate, and it is only recently with the application of statistical methods to the problem that some ideas of its fundamental nature are being uncovered. At present it is only possible to approximate the magnitude of the diffusion rate.

Sorption

Some of the radioactive salts introduced into an aqueous media tend to dissociate into their ionic forms. Once the radioactive material becomes cationic and anionic, it is more likely to be sorbed on the organic and clay particles suspended in the stream and on the river bottom. Clay materials have a particular affinity for cationic materials, and this includes most of the hazardous fission products.[3] In a series of experiments CARRITT and GOODGAL determined the percentage of sorption of P^{32}, I^{131}, Sr^{89}, Cu^{64}, Fe^{59} and S^{35} on Chesapeake Bay surface sediments (top 6 in. and 6 in. at 9 ft).[4] The composition of the sediments was approximately 5 per cent kaolinite, 30 per cent illite, 40 per cent chlorite-like montmorillonite, 15 per cent amorphous silica, and 10 per cent quartz.

For the conditions to be expected in the estuaries, the uptake of phosphorus was less in sea water than in fresh-water systems, but in both systems approached 100 per cent removal within 1 hr. Iodine showed no significant uptake, but most of the strontium which was not precipitated was sorbed on the muds in less than 30 min. It is reasonable to assume that an appreciable fraction of radiostrontium waste will become associated with naturally suspended solids. Copper was almost completely removed in less than 24 hr, but iron and sulfate were not removed at all.

In general, therefore, it may be said that alkali metals (group IA), alkaline earth metals (group IIA), and transition elements (group IIIA) in the periodic table are sorbed quite easily on the sediments, while elements of the transition groups—VIIA, VIII, IB, and IIB—the non-metals (group VIB) and the halogens

(group VIIB) are sorbed more slowly. These are only general conclusions. For specific chemical entities, the quantitative sorptions would depend upon the chemical characteristics of the radioactive material and the receiving waters, such as pH, temperature, concentration and ionic strength.

In addition to sorption on the muds, there can be considerable uptake and concentration in the biota. The organisms that sorb the isotopes from the water take the elements into their system to the degree required to satisfy that system. Therefore radioactive isotopes can be concentrated many times by particular organisms and in a particular part of the organism. There are wide variations in these factors between organisms, but approximate concentration factors are shown in Table 1.[5]

Sedimentation

The sedimentation of clay particles and dead organic matter will also remove the radioactive materials attached to the matter. Whenever any of the dissociated compounds exceed their solubility product, they will precipitate and settle out. In general, when fresh water enters salt water, some of the colloidally dispersed solids flocculate and settle.

The sedimentation process can be further increased by applying flocculating agents to the water. A group at the Armour Research Foundation has studied the possibility of such processes by using aluminum and ferric hydroxide systems.[6] Since the radioactive materials will disperse in the ocean quite rapidly, only a system that immediately operates upon the introduced contaminant can be effective.

Table 1. *Approximate concentration factors of different elements in members of the marine biosphere.*
(Concentration factors are based on a live weight basis.)

Element	Form in Seawater	Concentration in Seawater (μg/liter)	Concentration in Noncalcareous Algae (μg/liter)	Concentration Factors			
				Invertebrates		Vertebrates	
				Soft	Skeletal	Soft	Skeletal
Na	Ionic	10^7	1	0.5	0	0.07	1
K	Ionic	380,000	25	10	0	5	20
Cs	Ionic	0.5	1	10		10	
Ca	Ionic	400,000	10	10	1,000	1	200
Sr	Ionic	7,000	20	10	1,000	1	200
Zn	Ionic	10	100	5,000	1,000	1,000	30,000
Cu	Ionic	3	100	5,000	5,000	1,000	1,000
Fe	Particulate	10	20,000	10,000	100,000	1,000	5,000
Ni[a]	Ionic	2	500	200	200	100	
Mo	Ionic-Particulate	10	10	100		20	
V	?	1	1,000	100		20	
Ti	?	1	1,000	1,000		40	
Cr	?	0.05	300				
P	Ionic	70	10,000	10,000	10,000	40,000	2,000,000
S	Ionic	900,000	10	5	1	2	
I	Ionic	50	10,000	100	50	10	

[a]Data from T. Laevastu and T. G. Thompson, "The Determination and Occurrence of Nickel in Sea Water Marine Organisms and Sediments," J. du Cons. 21, 125-143 (1956).

All the systems described above interact with each other, and the efficiency and effectiveness of any one system is also a function of the others. For example, when the contaminant precipitates and settles, it comes into contact with other suspended solids and may sorb them and thus scavenge more material than the original precipitation.

Radioactive decay

The radioactive contaminants decay with time, which decreases the concentration of the isotopes in the water, muds and biota.

MODEL STUDIES

As outlined in the preceding section, the determination of the diffusion and convection coefficients in the field for an area the size of the Delaware River Basin is quite difficult. An easier solution is to build a physical model to represent the field conditions and to determine the diffusion and convection of a contaminant from a model. A large-scale model of the Delaware River was built in 1948 by the Corps of Engineers at the U.S. Army Waterways Experiment Station, Vicksburg, Mississippi. This model is based upon a 1942 hydrographic survey for the studies of salinity intrusions, shoaling, and the flushing of waste discharged into the estuary.[7] The model is 750 ft long and 130 ft wide at its widest point and covers an area of 30,000 ft^2, which is equivalent in the prototype to 100 miles^2. The general area is shown in Fig. 1 and the detailed area in Fig. 2. Because of the large area covered and the relatively shallow depths, it has been necessary to use a distorted model so that the depths of water in the model will be great enough to ignore surface tension.

The Delaware model is scaled 1:1000 horizontal and 1:100 vertical. It follows then from the Froude scaling ($v/\sqrt{gl}$) (where v is a velocity term, g is the gravitational acceleration, and l is a length term) that the velocity is scaled 1:10, time 1:100, discharge 1:1,000,000, and volume 1:100,000,000. The salinity scale ratio is 1:1.

Prior to using any model, it is necessary to make sure that the model is not only geometrically similar to the river, but that the velocity, tidal heights and elevations, and salinity profiles are also similar. The hydraulic agreement between the Delaware model and prototype is considered to be excellent. The prototype representation of tidal heights is accurate within $\frac{1}{10}$ ft, velocity within $\frac{5}{10}$ ft/sec and salinity within 5 per cent of values measured in the river.[8]

Model tests of 1952

A series of tests were run in the model in 1952 to determine the effects of a pollutant introduced into the Delaware estuary. In two of the tests, the contaminant was introduced into the main channel opposite the Philadelphia Naval Shipyard, Delaware River Mile (D.R.M.) 96.6. The pollutant was introduced at high-water slack in the center of the channel and samples taken at high-water slack from the center of the channel. During the first test methylene blue dye, used as the contaminant, was instantaneously released at mean tide and mean-river flow (12,350 ft^3/sec at Trenton) and during the second test at low-river flow (3000 ft^3/sec at Trenton). The results of the tests are shown in Figs. 3 (PRITCHARD[7], pp. 28–29) and 4 (PRITCHARD[7] p. 34).

This study is concerned with the effects of a release of a radioactive contaminant during start-up operations at the New York Shipbuilding Corporation outfitting docks 22,500 ft upstream (D.R.M. 101) from the dosing site. Since this is only about one-half of a tidal excursion upstream, it was felt that no serious error would be introduced by transposing the dosing site upstream.

The main difference in the results was caused by the fact that the Schuylkill River enters the Delaware River just below the original dosing point at the U.S. Naval Shipyard and thereby provides greater dilution and turbulence on the first ebbing of the tide after dosing.

Six cases have been studied: (1) the instantaneous dose, (2) equilibrium values of an instantaneous dose at each tidal cycle, (3) equilibrium values of an instantaneous dose every other tidal cycle (approximately once each day), at mean flow of 12,350 ft^3/sec at Trenton and at low-water flow of 3000 ft^3/sec at Trenton. The dosing conditions were not typical of those that might occur in the Camden outfitting basin, because the accident, if one occurs, would most likely occur at dockside, rather than the center of the stream. The resulting spread of contaminant would then be slower by a considerable amount because of lesser mixing and turbulence near the shore. The initial dosing also covered one-third of the width of the stream which facilitated the dispersion. All of these changes are on the conservative side. On the other side is the fact that the accident would most likely not occur directly on high-water slack and, therefore would occur under more turbulent conditions than those of the test. It is obvious from a study of the dye dispersion that the difference between discharge at high- and low-water slacks is inconsequential after one or two tidal cycles. The increased area in the cross section at high tide is only 20 per cent greater than the area at low tide. The concentration of the contaminant, however, decreases by a factor of 100 after just one tidal cycle, and by a

FIG. 1. Map of Lower Delaware River Valley.

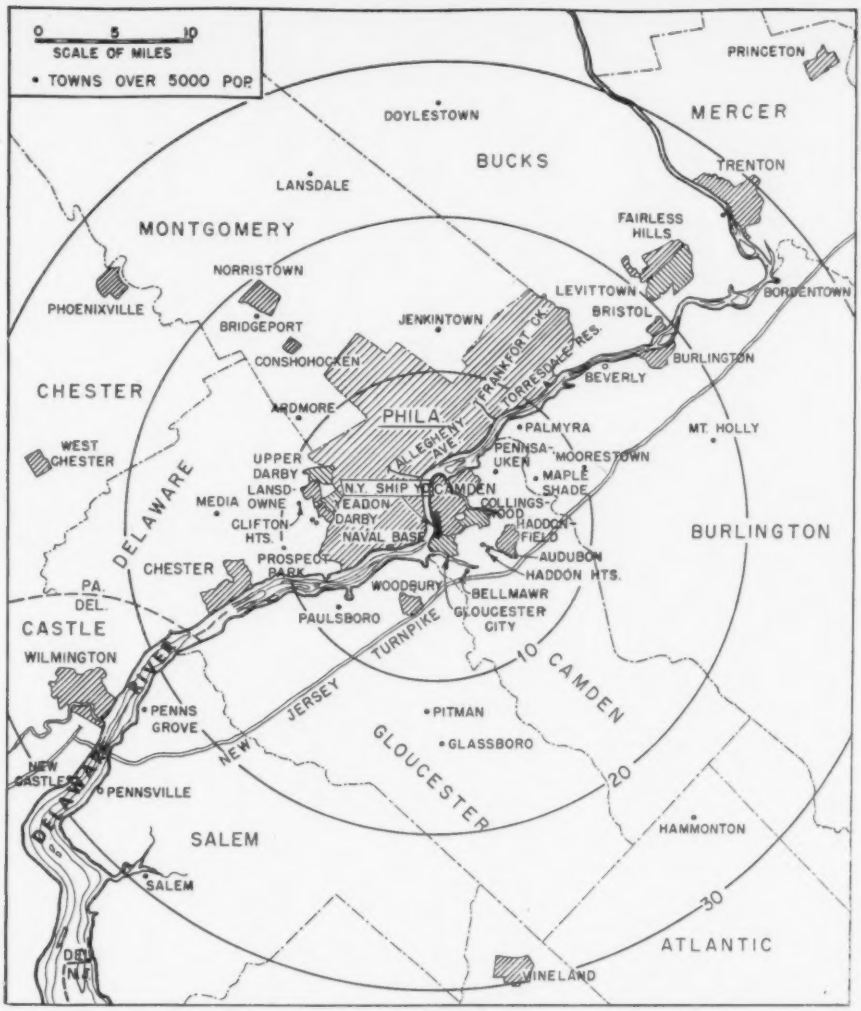


FIG. 2. Map of Philadelphia–Camden area.

factor of 1000 in three tidal cycles at the mean flow.

The results of the model study are applicable only to a conservative contaminant; that is, one in which there is no decay nor removal by any physical or biological mechanisms. The decrease in concentration is due only to the diffusion and dispersion in the liquid phase. A radioactive contaminant, on the other hand, is a non-conservative contaminant in that it is accumulated in destructive fashion in the river, and it decays with its own characteristic half-life in the sediments, flora and mud in the river. All these facts make the results based on the model study extremely conservative. This, of course, must be balanced against the other assumptions enumerated above, but, in general, it is possible to say that the results based on the model study are conservative and particularly so for the short half-life isotopes.

The most serious accident that could occur in the river would take place if the Torresdale Filter Plant (D.R.M. 114), which is about 13 miles above the New York Shipbuilding outfitting basin, and which supplies Philadelphia with about one-half its water, should become contaminated. Model studies indicate that this is not likely, but this is somewhat deceiving

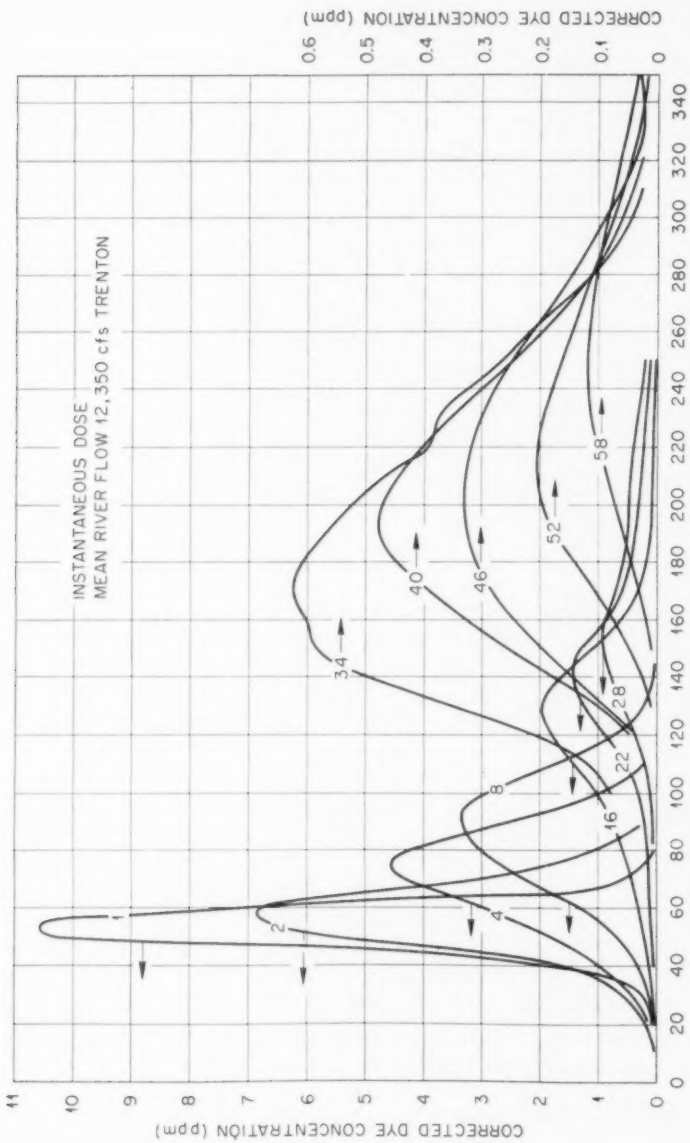


FIG. 3. Longitudinal distribution of contaminant after designated number of tidal cycles.

FIG. 4. Longitudinal distribution of contaminant after designated number of tidal cycles.

as the limits of detection of methylene blue, used as the contaminant in the model study, are far below the limits of detection of any radioactive isotopes. For example, the limit of detection of methylene blue, the dye used in model studies, was 0.01 p.p.m. or 0.01 mg/l, and 0.01 mg Sr^{90} per l. is equal to 1.45×10^{-3} c/l. (1.45×10^{7} times the MPC value of 1×10^{-10} c/l.) or at 10 per cent counting efficiency equal to 3.2×10^{5} counts/min per ml. The limit of detection of Sr^{90} unconcentrated is, of course, a function of the counting equipment and sample preparation, but we can say that from 10 to 15 counts/min per ml are easily detectable; there is, therefore, a factor of 10^{4} difference in the ease of detection. This large difference might be an excellent reason to consider the use of radioactive isotopes rather than dye in future tests.

Assumptions

In order to correlate the results of the model tests with the results from an accident, we assume that parts per million (p.p.m.) is equivalent to milligram per liter (mg/l.), as this is actually the way the parts per million are measured in most laboratories. If, as above, it is assumed that the dose in curies and the contaminant in millicuries per liter will be proportional to the quantity of dose in grams and milligrams per liter of contaminant, the maximum contamination for different accidents may be obtained. Correspondence between milligrams of dye and milligrams of isotopes and linearity of response from larger doses are the two important assumptions in using the model data.

Model tests of 1958

The Delaware River was subjected to a detailed hydrographic resurvey in 1954, and the model was revised accordingly.

At the request of the Nuclear Projects Office of the Maritime Administration, tests were run on the revised model for a 3 hr and a twenty-cycle dose at low-water slack at New York Shipbuilding outfitting basin during both mean and low flows of fresh water. In addition, a test was run in which half the dose was discharged in 3 hr at low-water slack and the remainder over twenty tide cycles at low-water flow.[9] The dosing was at low-water slack, as this would tend to give the highest concentrations of contaminant at Torresdale water intake. If dosing had been done at high-water slack, the contaminant would have been swept downstream prior to being swept upstream and thus would have been diluted and dispersed to a considerable extent prior to reaching Torresdale. The results of this last test are shown in Fig. 5 and compared with the results of the computed values from the 1952 tests and shown to be in fair agreement (Fig. 6). The earlier tests were used to approximate a continuous release by assuming an instantaneous release to occur once each tidal cycle. The increment of concentrations from each new addition was added to the (dispersed) accumulated concentrations from the previous additions.

Similar calculations can be made for any other section of the river or taken from the data of the second set of model experiments. These give the concentrations and the times at which they exist at any particular river station for a conservative contaminant. Based on the hypothesized accident, it is possible then to determine the distribution of isotopes in the river, assuming that they act as conservative contaminants.

Maximum credible accident

The hypothesized accident investigated is usually the maximum credible accident. The rupture of the containment vessel of the NS *Savannah* in a port, harbor, or river area is not a credible accident (COTTRELL *et al.*,[10] p. 36). Therefore, the maximum credible accident will assume leakage from the unruptured containment vessel.

The maximum credible accident assumes that the accident occurs after long-term operation. The fission-product inventory is that after 600 days operation at 69 MW. It is further assumed that 100 per cent of the fuel rods have failed and that the following percentages of the accumulated fission products are released to the containment vessel (COTTRELL *et al.*,[10] p. 16):

Element	Released (%)
Xe, Kr, Rb, I, Br, Cs	100
Sr, Ba	10
Te, Se, Sb, Eu, Mo, Tc, Sm	1
Y, La, Ce, Pr, Nd, Pm, Zr, Nb, Ru, Rh	0.1

The containment vessel is assumed to leak at a rate of 0.2 per cent of the free volume per day.

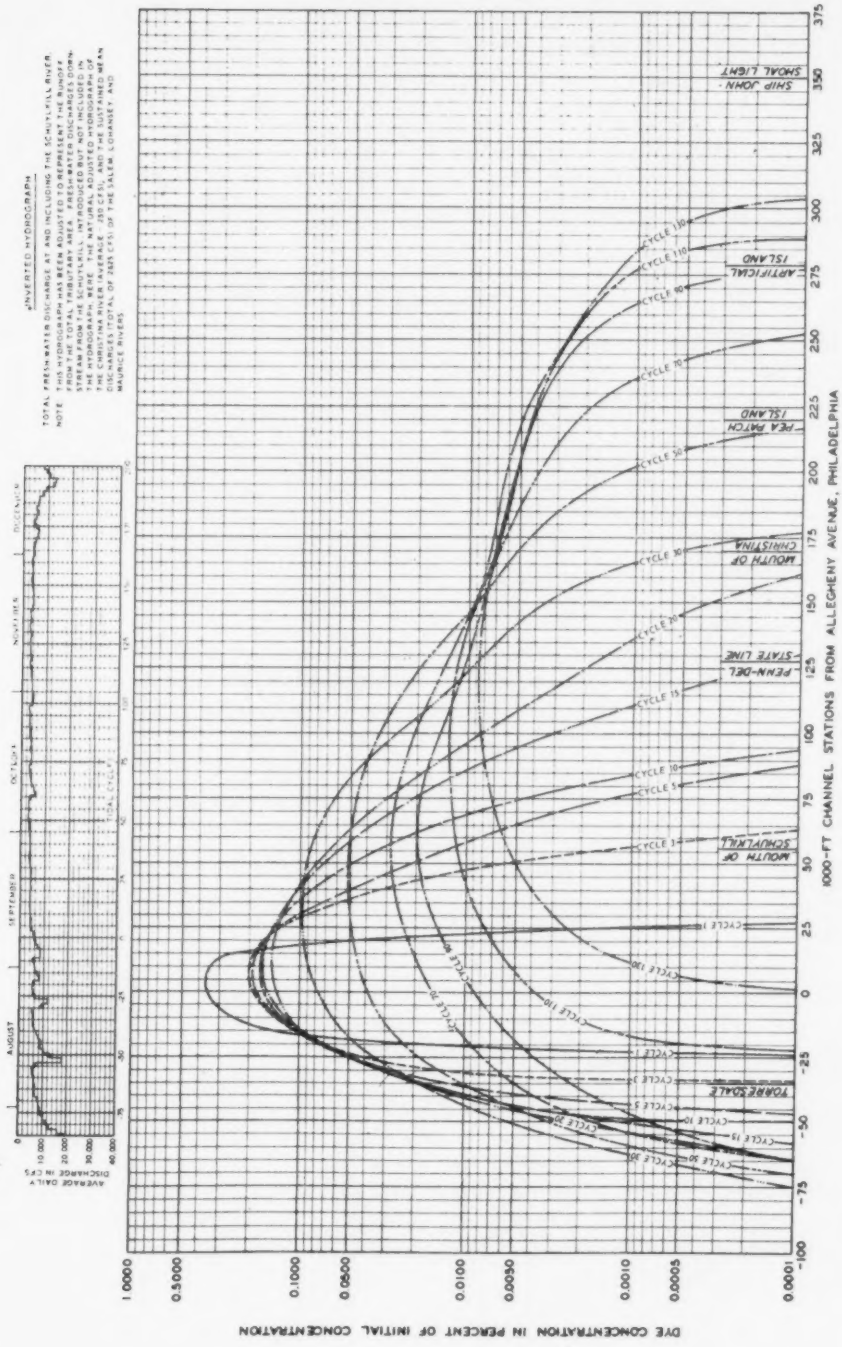


Fig. 5. Distribution of contaminant at highwater slack.

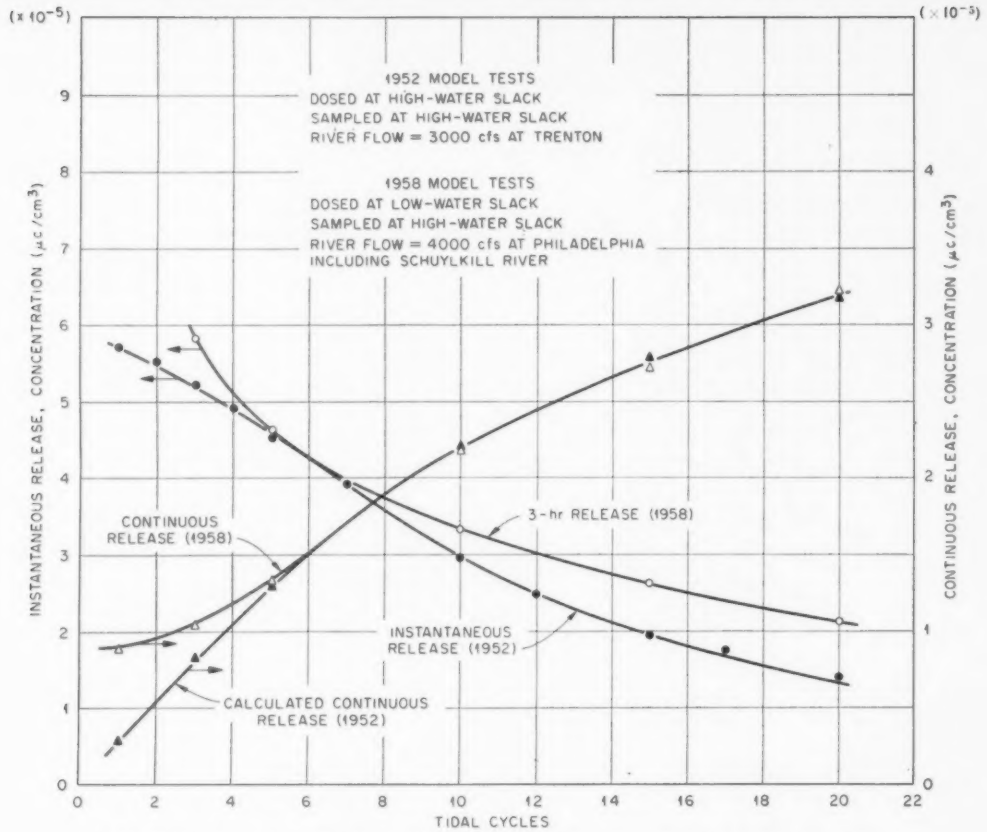


FIG. 6. Concentration at Benjamin Franklin bridge for dose of 10^4 c at NYS outfitting basin as a function of time in tidal cycles.

The main isotopes of interest that the containment vessel would contain are listed in Table 2 (COTTRELL *et al.*[10], p. 39).

Exposure calculations

The 1958 tests, 1 and 4, assumed an instantaneous release (3 hr) which is not a credible accident. Deposition of air-borne activity is approximately an instantaneous release and for the worst case results in deposited activity in the river four to five orders of magnitude less than that by direct release to the river (3.2 c vs. 6.2×10^5 c). (Ref. 10, pp. 40 and 54.)

Test 2, in which one-half the activity was released in 3 hr and the remainder during twenty tidal cycles, was judged to be most similar to any accident that might occur. This produced a maximum exposure, less than 60 per cent of the exposure produced by test 3, which was a uniform release over twenty tidal cycles at low-river flow and less than 70 per cent of the exposure produced in test 5, which was a uniform release over twenty tidal cycles at mean-river flow (as shown in Figs. 7–9 Ref. 10, pp. 46, 48 and 49, respectively).

The critical organ exposures were computed from the concentration of fission products in the river at a given location and at a given time as a percentage of drinking water MPC and submersion exposure in mrem/hr if there is swimming or boating on the river.

The percentage exposure of drinking water MPC was calculated for thyroid, bone, total

Table 2. Source data for isotopes of interest in hydrospheric release

Isotope	Half Life	Gamma Energy, E_γ (Mev/dis)	Activity Release to Containment Vessel Q_T (μc)	MPC Exposure (μc/cm^3) to Critical Organ from Drinking Water[a]			
				Thyroid	Bone	Total Body	GI (LLI)
Sr^{89}	54 d		2.57×10^{11}		10^{-4}	7×10^{-4}	4×10^{-4}
Sr^{90}	28 y		1.31×10^{10}		10^{-6}	4×10^{-6}	5×10^{-4}
Sr^{91}	9.7 h	0.85	3.2×10^{11}		7×10^{-3}		7×10^{-4}
Y^{91}	58 d		3.2×10^{9}				3×10^{-4}
Y^{93}	10.0 h	0.7	3.6×10^{9}				3×10^{-4}
Zr^{95}	63 d	0.73	3.8×10^{9}				6×10^{-4}
Zr^{97}	17.0 h	0.75	3.5×10^{9}				2×10^{-4}
Te^{129m}	33 d	0.106	2.07×10^{9}	8×10^{-3}	3×10^{-3}	2×10^{-3}	3×10^{-4}
Te^{132}	77 h	0.231	2.57×10^{10}			5×10^{-3}	3×10^{-4}
I^{131}	8.05 d	0.39	1.97×10^{12}	3×10^{-5}[b]		2×10^{-3}	(Insoluble[c]) 6×10^{-4}
I^{132}	2.4 h	1.99	2.57×10^{12}	8×10^{-4}[b]		4×10^{-2}	
I^{133}	20.8 h	0.56	3.7×10^{12}	10^{-4}[b]		9×10^{-3}	(Insoluble[c]) 4×10^{-4}
I^{134}	52.5 m	1.27	4.3×10^{12}	2×10^{-3}[b]			
I^{135}	6.68 h	1.54	3.4×10^{12}	4×10^{-4}[b]		2×10^{-2}	(Insoluble[c]) 7×10^{-4}
Cs^{136}	13 d	1.02	3.7×10^{10}		10^{-2}	9×10^{-4}	(Insoluble[c]) 6×10^{-4}
Cs^{137}	26.6 y		1.15×10^{11}		5×10^{-4}	2×10^{-4}	(Insoluble[c]) 4×10^{-4}
Ba^{140}	12.8 d	0.237	3.6×10^{11}		2×10^{-3}	5×10^{-3}	3×10^{-4}
La^{140}	40.2 h	2.11	3.5×10^{9}				2×10^{-4}
Ce^{144}	290 d	0.043	2.57×10^{9}				10^{-4}
Pr^{143}	13.7 d		3.5×10^{9}				5×10^{-4}
Mo^{99}	67 h	0.24	3.5×10^{10}			8×10^{-3}	2×10^{-3} (Insoluble[c]) 4×10^{-4}
Kr^{83m}	114 m	0.0415	2.57×10^{11}				
Kr^{85m}	4.36 h	0.181	7.8×10^{11}				
Kr^{87}	78 m	0.56	1.43×10^{12}				
Kr^{88}	2.77 h	2.07	1.97×10^{12}				
Xe^{133m}	2.3 d	0.233	7.3×10^{10}				
Xe^{133}	5.27 d	0.081	3.7×10^{12}				
Xe^{135m}	15.6 m	0.52	1.02×10^{12}				
Xe^{135}	9.13 h	0.268	3.5×10^{12}				
		Total	2.99×10^{13}				

[a] Based on permissible occupational exposure for 168-hr week.

[b] These values were taken from NBS Handbook 52. This has now been superceded by NBS Handbook 69, in which the thyroid MPC's for $I^{131-135}$ are listed as 2×10^{-5}, 6×10^{-4}, 7×10^{-5}, 1×10^{-3} and 2×10^{-4}, respectively. The differences that corrections to the later list would make in the pertinent tables and graphs are small.

[c] Not included in the calculations.

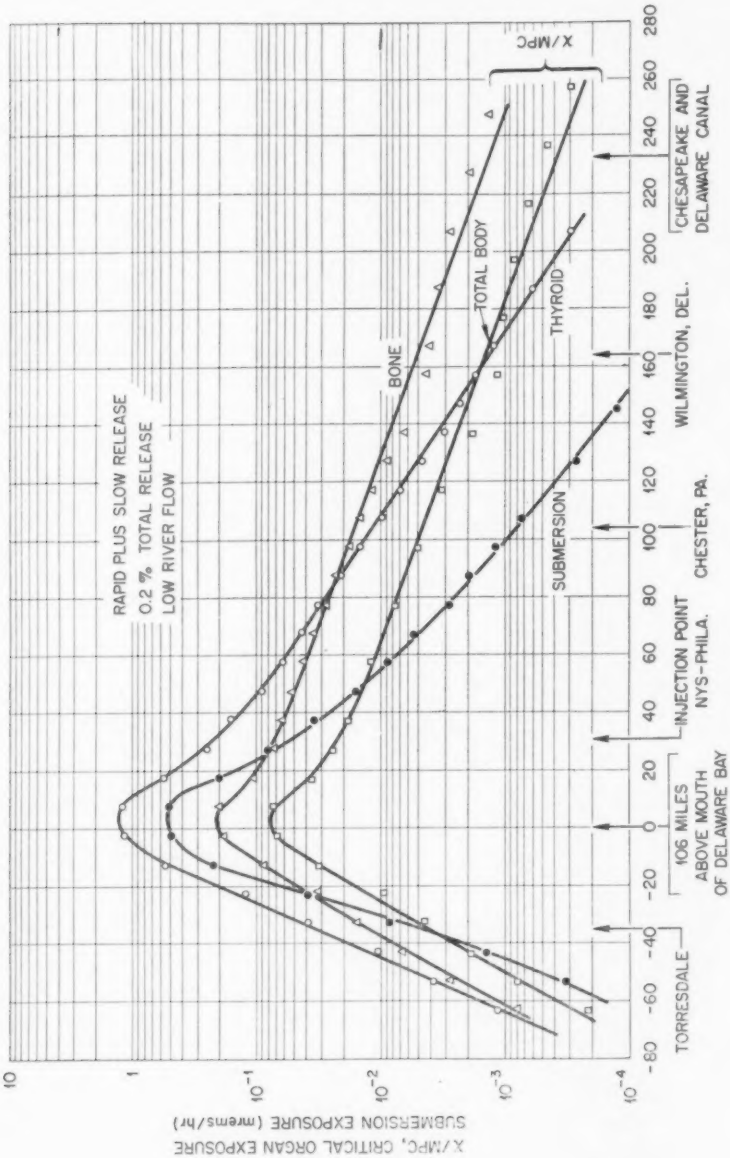


FIG. 7. Maximum exposure as a function of river location (model test 2).

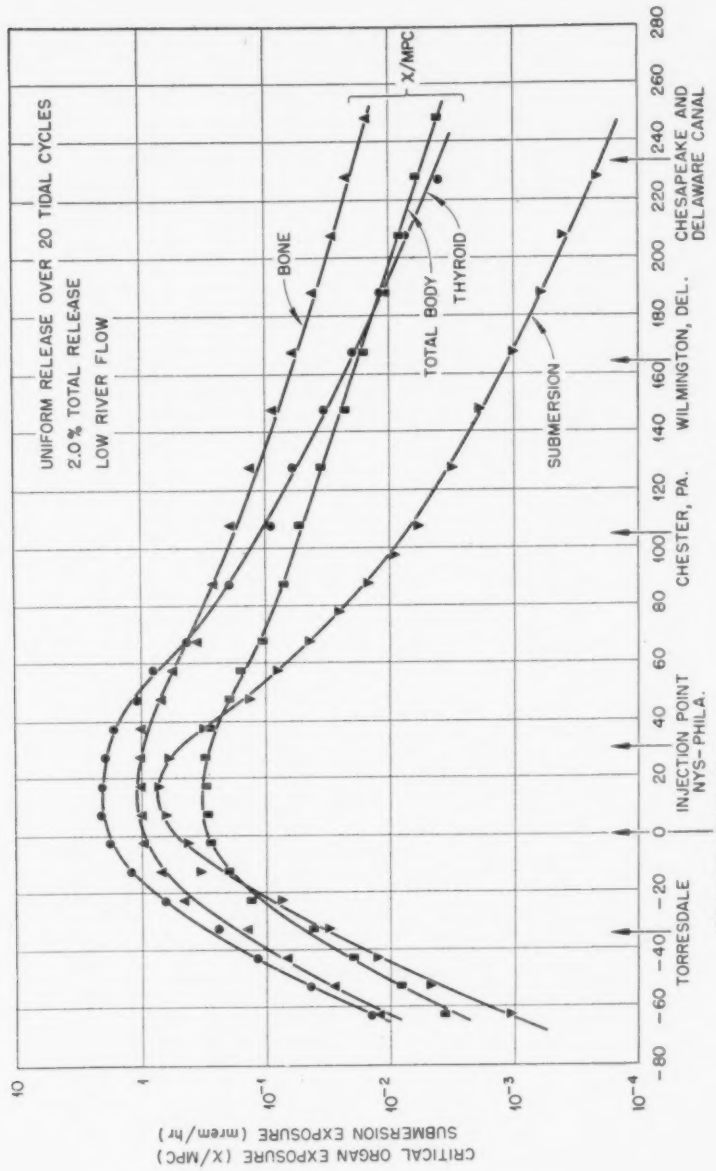


FIG. 8. Maximum exposure as a function of river location (model test 3).

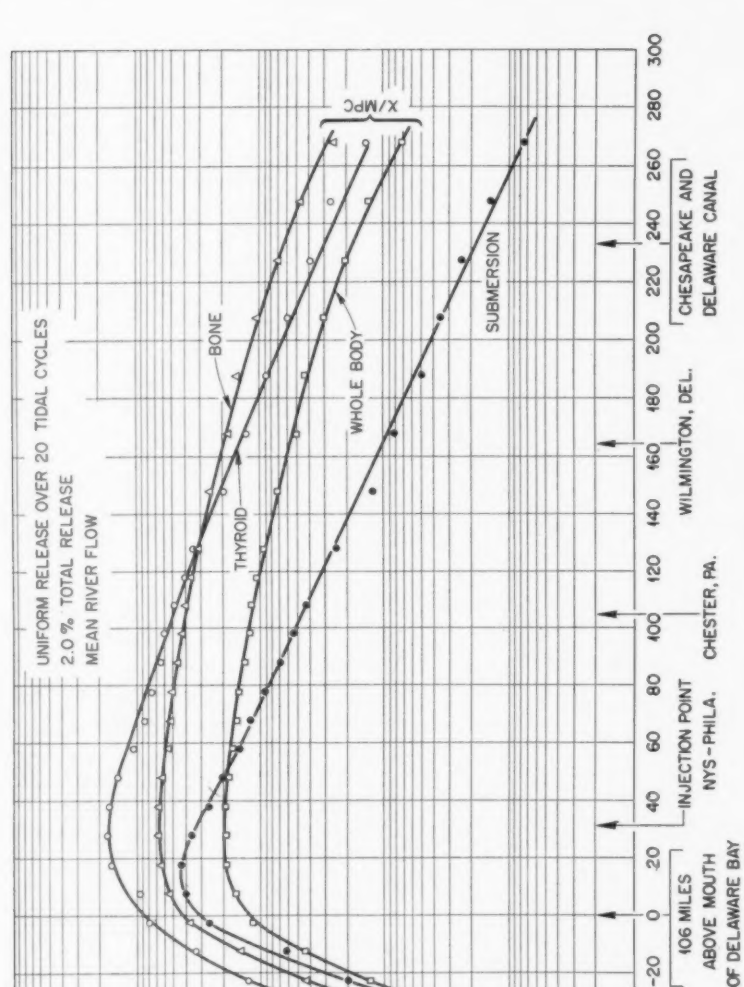


FIG. 9. Maximum exposure as a function of river location (model test 5).

body and gastrointestinal tract (lower large intestines).

The fraction of the MPC value was calculated from equation (2):

$$\sum_i \left[\frac{Q_i(t)}{\mathrm{MPC}_i}\right]_{co} X/Q \qquad (2)$$

where $Q_i(t)$ = source strength of isotope i (μc);
MPC_i = MPC of isotope i(in μc/cm^3) for water;
X/Q = dispersion value (in cm^{-3}) obtained from model data;
co = critical organ.

The dose rate from submergence in an infinite volume of water is given in equation (3):

$$D = \int_0^{4\pi} C(E)\,\frac{S e^{-\mu(E)R} B_r(\mu R)\, d\Omega}{4\pi R^2}, \qquad (3)$$

where D = dose rate (rems/hr);
$d\Omega$ = the differential solid angle (steradians);
$C(E)$ = flux-to-dose rate conversion factor [(r/hr)/MeV/cm^3 sec];
S = source strength (MeV/cm^3 sec);
$B_r(\mu R)$ = dose build-up factor for point isotropic source (dimensionless);
$e^{-\mu(E)R}$ = total attenuation factor (dimensionless);
μ = attenuation factor per unit length (cm^{-1});
R = straight-line path length (cm).

$$S = 3.7 \times 10^{10} \sum_i Q_{T,i} E_{\gamma,i} \frac{X}{Q} \frac{Q_0}{Q_T} \qquad (4)$$

where $Q_{T\,i}$ = total amount of isotope i (c) released to containment vessel for failure of 100 per cent of the fuel elements;
$E_{\gamma,i}$ = γ-energy of isotope i (MeV/dis);
X/Q = dispersion (cm^{-3}).

The summation of $Q_T E_\gamma$ is shown on Fig. 10, and by using the value Q_0/Q_T, where Q_0 is the total curie release to the river and Q_T is the total curie release to the containment vessel, for the particular test, and time and X/Q values

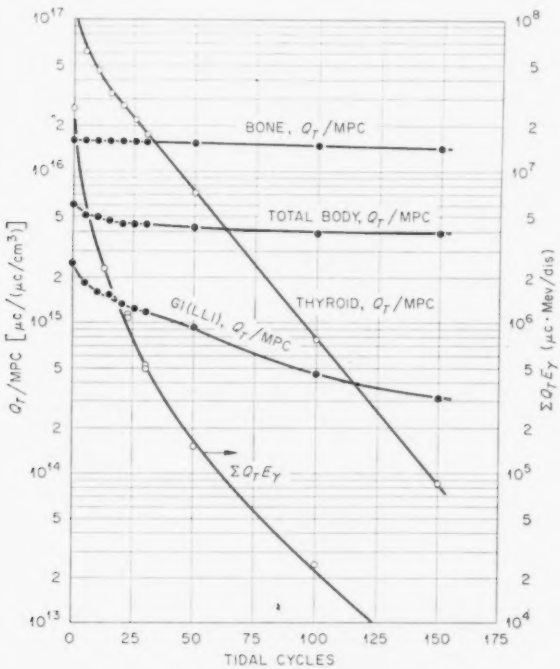


FIG. 10. Critical organ and submersion dose as a function of time in tidal cycles.

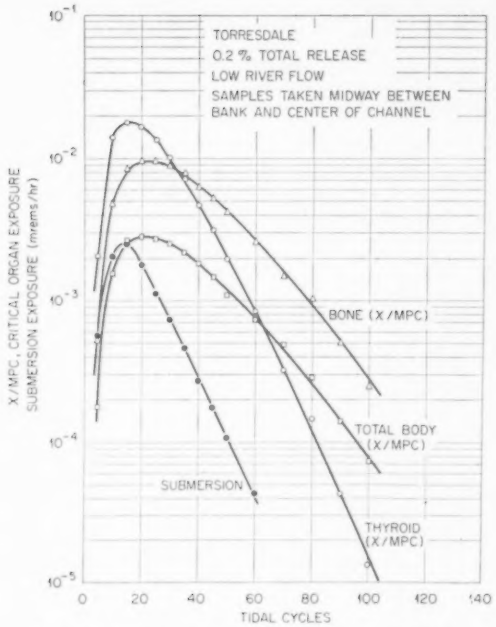


FIG. 11. Exposure at Torresdale Municipal Water Intake Plant as a function of time in tidal cycles (model test 2).

from the model tests, the submersion dose can be calculated. The maximum submersion doses are shown on Figs. 7–9. It is of equal interest to determine the time history of the exposure at the most critical site, the Torresdale Municipal Water Intake Plant as shown in Fig. 11. The results were obtained using the above equations and the suitable decay factors from Fig. 10 and dispersion factors from Ref. (9). Similar calculations have been carried out for stations at Wilmington and Philadelphia and could be done for any of the other stations in the model.

Sedimentation and sorption

These studies in the model modified by the decay calculations show the effects of convection, diffusion and radioactive decay. The other two major processes effecting the dispersion of radioactive material in the river are sorption and sedimentation, which must be taken into account as the contaminant is non-conservative. The sedimentation and sorption coefficients are extremely uncertain and difficult to predict. They vary widely from species to species and from isotope to isotope. The problems are so numerous that the Committee on Oceanography of the National Academy of Sciences—National Research Council stated:

> In arriving at recommended disposal rates the interaction of a contaminant with suspended solids and bottom sediments has been neglected. It was found impossible to make a quantitative estimate of the magnitude of this reaction. Neglecting this factor puts a certain factor of safety in the recommendations, as sorption onto bottom sediments within the disposal area will provide additional containment, thus allowing for further destruction of the contaminant by radioactive decay.[11]

However, for the purpose of this study, it was felt that these effects should be evaluated with the best data presently available. It is recognized that the values used are not definitive and may be in error by orders of magnitude. However, an estimate of the effects can be made once some values are assigned to the

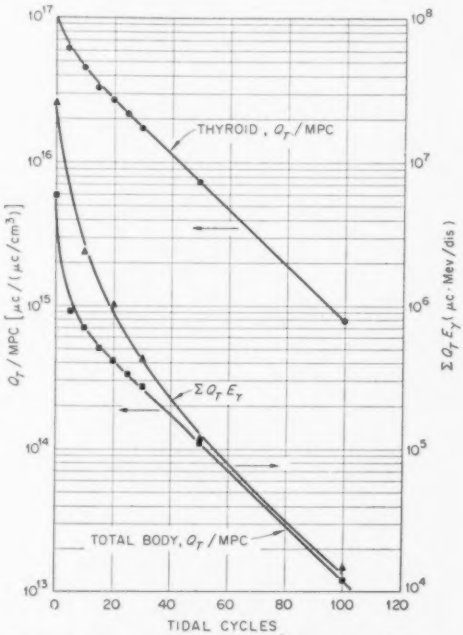


FIG. 12. Critical organ and submersion dose as a function of time in tidal cycles, including uptake.

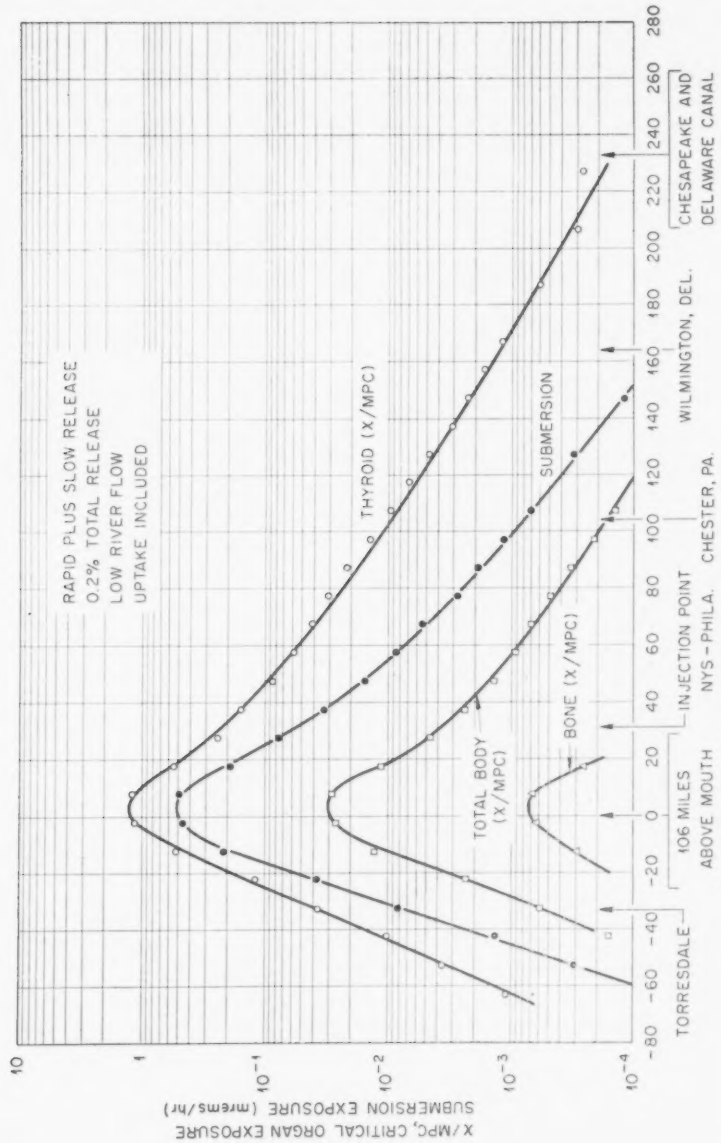


FIG. 13. Maximum exposure as a function of river location with uptake (model test 2).

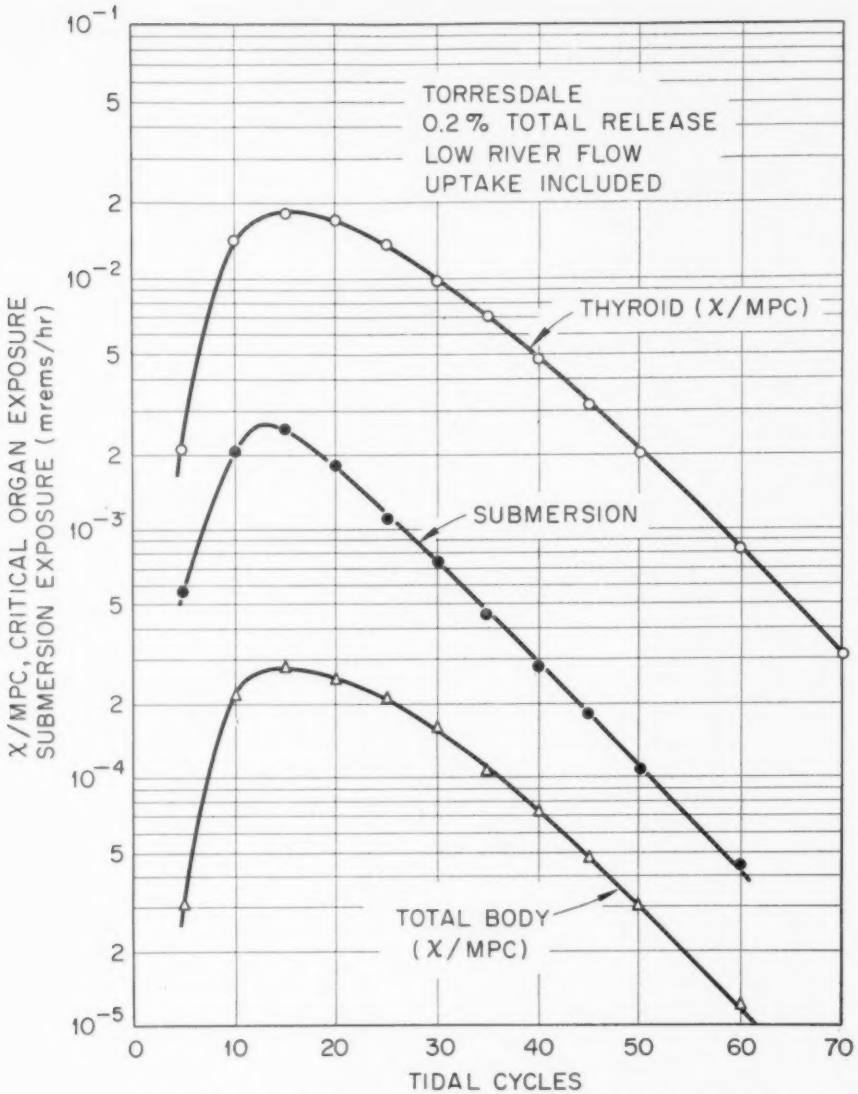


FIG. 14. Exposure at Torresdale Municipal Water Plant Intake as a function of time in tidal cycles with uptake (model test 2).

sorption and sedimentation coefficients. Since they are both so indefinite a combined coefficient λ (hr^{-1}) was used; for I^{131}, $\lambda = 0$; for Sr^{90}, Cs^{137} and Ce^{144}, $\lambda = 0.7$; and all other isotopes, $\lambda = 0.1$. The values chosen are based upon the works of CARRITT and GOODGAL[4], KETCHUM and BOWEN[12], and PENDLETON and HANSON[13]. Since the sorption and sedimentation are to some extent concentration dependent, it was felt that an exponential decay would best express mathematically what was taking place physically and would parallel reduction in concentration caused by radioactive decay and eddy diffusion and convection.

Taking these factors into account as shown in Fig. 12 and applying them to test 2, the most typical case, it is obvious from Fig. 13 that iodine is the major hazard at all stations, since it is essentially unaffected by sorption and sedimentation. Similar effects are found at Torresdale Water Plant Intake for the time history of exposure as shown in Fig. 14.

CONCLUSIONS

From the data herein reported, it is seen that even the maximum credible accident to NS *Savannah* at Camden, New Jersey, and the subsequent discharge of fission products to the hydrosphere will not result in unacceptable exposures. Such an accident will at most cause a temporary maximum concentration in the river water twice the maximum permissible concentration for drinking water for continuous occupational exposure and a submersion dose of 0.7 mrems/hr. Though estuaries can be classified in major types, each estuary harbor and port must be evaluated separately to determine the exposure that will be caused by a maximum credible accident.

REFERENCES

1. F. L. Parker, *Trans. Amer. Geophys. Un.* **39,** 3, 434 (1958).
2. H. U. Sverdrup, M. W. Johnson and R. H. Fleming, *The Oceans, Their Physics, Chemistry, and General Biology* p. 91. Prentice-Hall, New York (1942).
3. K. E. Cowser and F. L. Parker, *Health Phys.* **1,** 152 (1959).
4. D. E. Carritt and S. H. Goodgal, *The Sorptive and Zeolitic Properties of Natural Water Borne Silts . . . in Natural Waters.* NYO-4591 (1953).
5. L. A. Krumholz, E. D. Goldberg and H. A. Boroughs, *The Effects of Atomic Radiation on Oceanography and Fisheries* Chap. 7, p. 74. National Academy of Sciences—National Research Council, Publication No. 551 (1957).
6. J. Rosinski, *Scavenging of Particulate Matter in Connection with Nuclear-Powered Ships.* ARF Project, C-119, Report No. 6 (1959).
7. D. W. Pritchard, *A Study of Flushing in the Delaware Model, Chesapeake Bay Institute.* The Johns Hopkins University Tech. Rep. VII (1954).
8. *Delaware River Model Study; Report* 1, *Hydraulic and Salinity Verification,* Technical Memorandum 2-337, U.S. Army Engineer Waterways Experiment Station, p. 23–24 (1956).
9. *Contamination Dispersion in Estuaries, Delaware River, Hydraulic Model Investigation,* Miscellaneous Paper No. 2-332, Report No. 1, U.S. Army Waterways Experiment Station (1959).
10. W. B. Cottrell, L. A. Mann, F. L. Parker and G. D. Schmidt, *Environmental Analysis of NS "Savannah" Operation at Camden,* ORNL-2867 (1960).
11. *Radioactive Waste Disposal into Atlantic and Gulf Coastal Waters,* Publication 655, National Academy of Sciences—National Research Council, p. 2 (1959).
12. B. H. Ketchum and V.T. Bowen, Biological factors determining the distribution of radioisotopes in the sea. *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva,* 1958, 15/P/402. United Nations, New York (1958).
13. R. C. Pendleton and W. C. Hanson, Absorption of cesium-137 by components of an aquatic community. *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva,* 1958, 15/P/392. United Nations, New York (1958).

Health Physics Pergamon Press 1961. Vol. 6, pp. 86–93. Printed in Northern Ireland

PROBLEMS OF PERSONNEL MONITORING AT A CRITICALITY ACCIDENT*

H. F. HENRY

Union Carbide Nuclear Company, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tennessee

(*Received* 15 *November* 1960)

Abstract—With respect to personnel monitoring at a criticality incident, the implications of some experimental data are reviewed which show the dependence of the relative positions of an individual, his typical personnel monitoring device, and the excursion which is essentially a unidirectional γ–neutron radiation source. These data show that, for a given exposure, the dose indicated by either a γ- or neutron-sensitive device of the type generally used can vary by factors of at least 3 or 4, due only to the attenuation by the individual's body. Similar data indicate qualitatively the effect of moderation and reflection in affecting the neutron spectrum incident upon the device as compared to that actually incident upon the individual. In view of the uncertainties thus indicated, it is concluded that, although a γ- and neutron-sensitive personnel monitoring device can provide sufficiently accurate data concerning an individual's exposure for record purposes and will be useful for the prompt identification of personnel who have been highly exposed, a monitoring system consisting of blood sodium determinations as calibrated by area type dosimeters is necessary to provide data which is useful in indicating medical treatment.

In view of the fact that many people consider personnel monitoring of radiation exposure at a criticality incident as merely an extension of routine health physics monitoring procedures, this paper will indicate some of the possible uncertainties inherent in the use of any simple personnel monitoring device, such as a film badge or similar item, where it is necessary to make accurate and prompt dose determinations for those who have been seriously exposed in a criticality incident and for whom careful medical attention, including hospitalization, will be indicated.[1] In general, this will include those who have received exposures of approximately 100 rads or more. Of particular importance would be such determinations where "heroic" measures for life-saving may depend upon the dose evaluation.

The principal reason that routine monitoring methods can not, in general, give data of the necessary accuracy is the fact that the radiation from an accident will be unidirectional whereas in routine monitoring activities, the incident radiation is usually multidirectional, thus permitting an "averaging" of the exposure with a consequent more accurate determination of the relative dose for various monitoring periods. Since such exposures are usually low, and the data obtained are primarily necessary for long-term record maintenance, exposures of this type will not be considered further in this article.

It may be noted that the dangerous short-term exposures resulting from an accident will generally occur during the reaction itself and will thus involve both neutron and γ-exposures; accordingly, the succeeding analyses will be concerned with uncertainties inherent in monitoring both radiation types.

The information presented concerning some of the uncertainties in simple neutron activation of indium foils was obtained by analyzing raw data obtained in 1950 and 1955, respectively, for use in calibrating the indium foil used in the ORGDP and Y-12 security badges.[2,3] Fig. 1 shows this security badge which proved of

* This document is based on work performed at the Oak Ridge gaseous diffusion plant operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

FIG. 1. Cutaway of security badge, showing indium foil.

BLANK
PAGE

great value in identifying those who were seriously exposed at the Y-12 incident in June 1958.[4] Fig. 2 shows the ORGDP calibration method using a critical uranium solution as has been previously described.[2,5] It will be noted that both bare indium foils and such foils enclosed within cadmium sandwiches were interspersed in paraffin block "phantoms"; essentially, this resulted in the determination of the thermal and indium resonant neutron fluxes as transmitted through the various thicknesses of paraffin.

The Y-12 method[3] is shown in Fig. 3 where the neutrons were obtained from "Godiva",[6] and the phantoms used were 1-ft^2 cans of sucrose–urea solution in proportions giving the same average chemical composition as the human body. It will be noted that the indium was placed on all four sides of these phantoms.*

No particular effort was made in either set of experiments to place the detectors so as to eliminate scattering from the surroundings and similar random effects. In fact, some of the units were placed near material with good moderating and reflecting properties to enhance these effects since it was assumed at the time of planning these experiments that such factors would simulate actual conditions, although it was recognized that the results would be somewhat complicated thereby.

Fig. 4, which is taken from the ORGDP data, indicates that an overall attenuation factor of approximately 3 has been attained with 8 in. of paraffin. This also reflects some effects of moderation in changing the neutron spectrum as will be described later. Fig. 5 gives similar information from the Y-12 data where the comparison is between an indium foil placed in front of and one behind a phantom, and the attenuation factor is noted to be of the order of from 3 to 7 for about 12 in. of solution.

Fig. 6 summarizes some data on γ-attenuation as measured at the ORGDP[7] along with some points obtained by BOND[8] and KINGSBURY[9]. It will be noticed that again

* In these experiments, three tests were made with "Godiva" inside of a building and a fourth one with "Godiva" outside of the building. These conditions are indicated in Figs. 5, 9 and 10 by the notation "Inside" and "Outside".

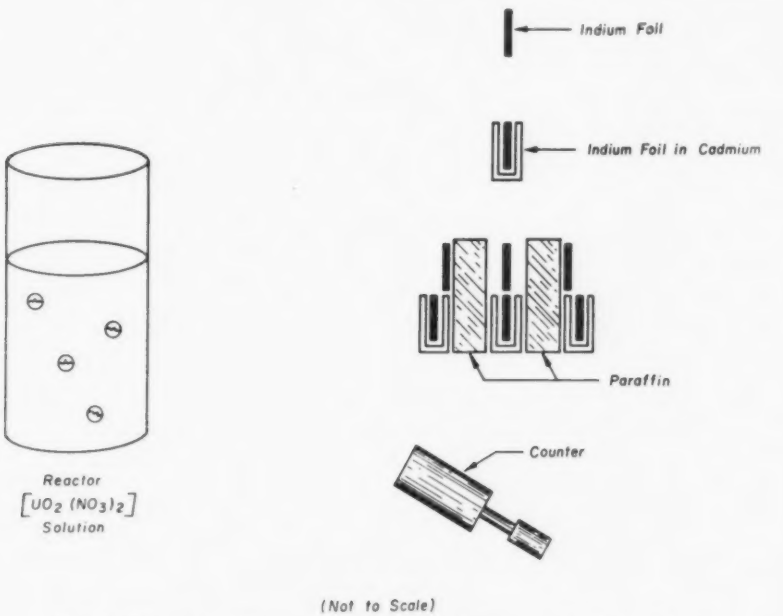


FIG. 2. Experimental arrangement Oak Ridge gaseous diffusion plant.

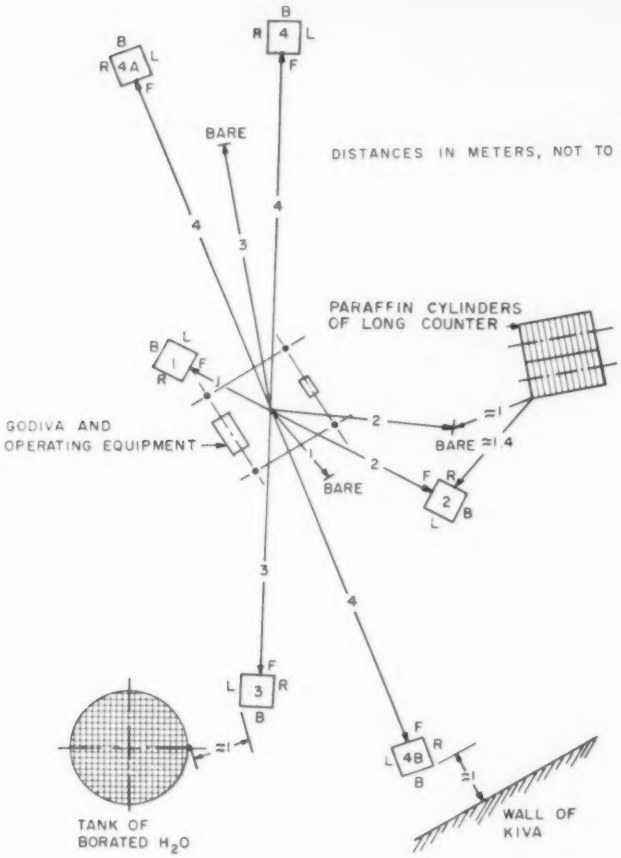
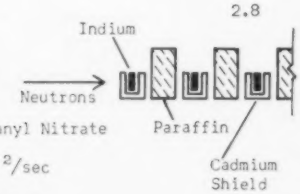


FIG. 3. Experimental arrangement of phantoms and badges Y-12 plant.

FIG. 4. Activations of indium foils* behind paraffin phantoms.

Paraffin Thickness (cm)	Activation (dis /min /g)	Activation at 0 / Activation at T
0	15,800	1.0
3.8	9,750	1.6
7.6	7,280	2.2
11.9	6,880	2.3
15.2	8,060	2.0
19.1	5,680	2.8



* Cadmium Covered - Data from K-736

Neutron Source - Aqueous Solution of Uranyl Nitrate

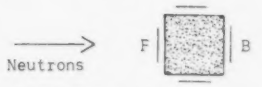
Leakage Neutron Flux = 3.435×10^4 n/cm ^{2}/sec

Distance from Reactor to Front Foil = 142 cm

Exposure Time = 108 min

Distance - Reactor To Phantom Front (Meters)	Inside Burst 1	Inside Burst 2	Inside Constant Power	Outside Constant Power
1	6.55	7.30	7.17	8.26**
2	3.00	2.57	2.57	11.72
3	1.94	2.26	1.95	9.80
4	2.53	2.81	3.13	13.20

* mr/hr /10^{17} fissions



12" Phantom - Aqueous Sol. of Sucrose and Urea

** Probably Low - Front Foil Activation Uncertain

Data from Y-1092

FIG. 5. Front foil activation*/back foil activation*.

attenuation factors of about 5 are indicated for a thickness of approximately 12 in.

In all of the above data analyses, the results indicate the discrepancy in the exposure indicated for an individual who is wearing a personnel monitoring device, such as a film badge, conventionally, when the reaction, which is then the source of radiation, occurs behind him. It is obvious that, due to the effects of attenuation, the relative positions of an individual, the detector, and a reaction would need to be very accurately known before the actual dose could be determined for a given exposure; without such information, uncertainties in exposure indications by factors of the order of 3 are easily possible for either neutron or γ-exposures.

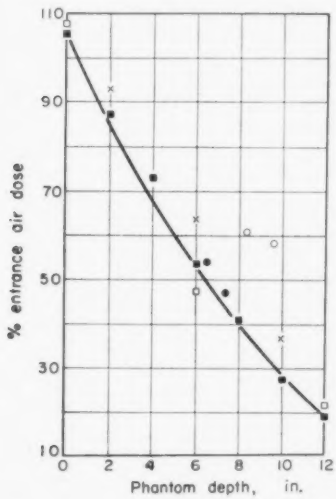


FIG. 6. Attenuation of γ-rays corrected from distance fall-off.

□■ Aqueous sucrose–urea; × masonite (BOND[8]); ○ human chest and ● human waist (KINGSBURY[9]). The square ■ at (0,105) covers a cross ×.

The use of a neutron detector also involves considerations of moderation and reflection by an individual's body with the result that the neutron flux spectrum affecting the detector will not be identical with that incident upon the individual.

Some of the effects of moderation and reflection are indicated by Figs. 7 and 8, which give the relative activations of bare and cadmium-covered foils in the ORGDP experiments. If it is assumed that practically all of the activation results from thermal and indium resonance neutrons, it may be assumed that only the resonance neutrons will affect a cadmium-sandwiched foil, but that both resonance and thermal neutrons will affect a bare foil.

Fig. 7 thus indicates that the foil activation due to the incident neutron beam used was due about equally to these two neutron energies. Fig. 8 shows the results of similar data taken with the foils placed in front of a paraffin block. In this case, it is apparent that both the bare and cadmium-sandwiched foils will be activated by incident and reflected neutrons. The cadmium-covered foils are activated by the

(Saturated Activity - dis /min /g)

Foil Set Number	Cd Shielded Foils	Bare Foils	Shielded* / Bare (%)
1	23,832	66,493	35.8
2	71,036	149,588	47.5
3	256,281	401,747	63.8
4	344,806	617,228	55.9
5	31,636	81,107	39.0
6	97,042	187,627	51.7
7	337,682	623,065	54.2
8	475,578	899,443	52.9
			Avg. 50.1

* Activity from resonant neutrons/activity from resonant and thermal neutrons

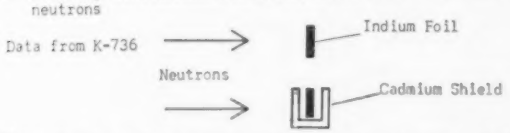


FIG. 7. Contribution of resonant and thermal neutrons to indium foil activations.

[Indium Foil Activity (Saturated) - dis /min /g]

Indium Foil Conditions	Foil Set #1	Foil Set #2	Foil Set #3	Foil Set #4
A. Bare	15,301	13,607	1,794	8,884
B. Cd Shield (on 1 side)	10,638	9,938	1,194	4,389
C. Cd Shield (on each side)	6,910	3,428	600	1,681
D. A.-C.	8,391	10,179	1,194	7,203
E. $\frac{A.-C.}{C.}$	1.21	2.97	1.99	4.28

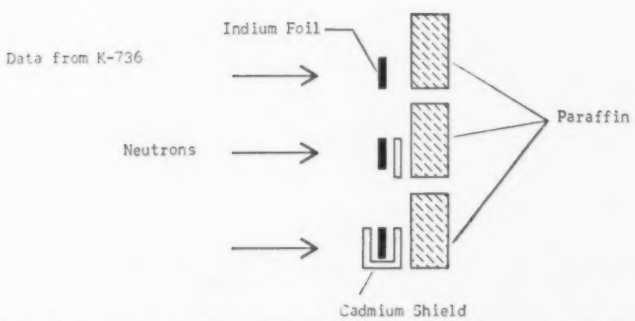


FIG. 8. Neutron moderation and back-scatter from paraffin reflectors.

Distance - Reactor To Phantom Front (Meters)	Inside Burst 1	Inside Burst 2	Inside Constant Power	Outside Constant Power	Outside/Inside Constant Power
1	6.17	7.36	7.37	-	-
2	3.24	2.85	2.84	4.70	1.65
3	3.06	2.50	2.37	5.08	2.14

* mr/hr /10^{17} fissions

Data from Y-1092

FIG. 9. Front foil activation*/"bare" foil activation*.

Distance - Reactor To Phantom Front (Meters)	Inside Burst 1	Inside Burst 2	Inside Constant Power	Outside Constant Power	Outside/Inside Constant Power
1	1.06	0.99	0.97	1.12	1.15
2	0.92	0.90	0.90	2.49	2.77
3	0.63	0.95	0.82	1.93	2.35

* mr/hr /10^{17} fissions

Data from Y-1092

FIG. 10. "Bare" foil activation*/back foil activation*.

resonance neutrons only, while the bare foils will be activated by both thermal and resonance neutrons. Hence, the data indicated on line *E* of this figure are the ratio of the activations due, respectively, to the thermal and resonance neutrons, both incident and reflected. It will be noted that the ratio is not unity as would be the case had there been neither selective reflection nor moderation, nor both, by the paraffin. With respect to practical monitoring, these data indicate that even when a neutron indicator is in the best possible position with respect to the reaction, the exposure which it indicates will differ from that actually incident on the individual since the incident neutron spectrum will be altered.

In general, evaluation of radiation scatter from surroundings is not particularly important since such scatter is also incident on the individuals concerned. However, some of the Y-12 data as summarized in Fig. 9 indicate that neutron back-scatter can produce activation which is as much as four times that due to the input incident radiation. Similarly, Fig. 10 indicates that forward-scatter, which is probably approximately the same as back-scatter, can be about the same as attenuation. Other data indicate that the effect of side-scatter is somewhat less than that of back-scatter. The effect of γ-scatter is only about 5–10 per cent and is thus negligible in comparison to other effects.[7]

Another factor which may affect the exposure indicated by personnel monitoring is the difference in the neutron and γ-spectra from

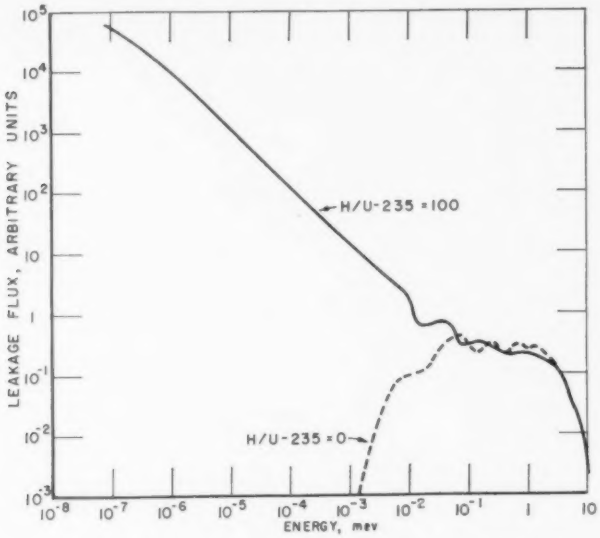


FIG. 11. Neutron leakage spectra for bare critical systems.

reactions in differing materials. Fig. 11 summarizes some calculations showing the effect of solution concentration on the neutron spectrum.[10] It has also been shown that the fraction of escaping neutrons which are at low energies varies from zero at fission to about 20 per cent in a well-moderated system, and the average dose per neutron does not vary markedly with moderation.[10] The γ–neutron dose ratio is probably even more important, and there have been indications that this dose ratio can be of the order of from 4:1 to 5:1 for well-moderated solutions, 1:1 for dry salts, and about 1:5 to 1:10 for metal systems.

Based on the data presented, it appears that the addition of a simple neutron-detecting foil to any presently used personnel monitoring device, or the use of such a foil independently, will make possible the prompt identification of employees who may have been exposed in an accident. This identification is extremely important for those who may have been sufficiently highly exposed that medical attention is necessary. Probably the simplest device is an indium foil although it may be noted that after a period of about 6 hr following an excursion, a gold foil will show a higher activity than an equal mass of indium. This device alone can give an indication of the actual exposure, especially where some information concerning the incident spectrum is available, but more complete data should be obtained if other γ and neutron exposure-indicating devices, such as those which might be employed in a normal personnel monitoring program, are also used. This may include various foil and film components with carefully chosen filters. Although the exposures indicated by these devices will be subject to the various recognized uncertainties inherent in any system of routine personnel monitoring, the necessary exposure data for inclusion in the personnel exposure records of the individuals concerned should be readily obtainable.

However, for high-level exposures for which medical decisions concerning specialized therapy may be promptly necessary, the probable uncertainties in dose determination due to orientation effects as described are sufficient to indicate the use of a type of measurement in which these uncertainties may be minimized. In this case, it appears that neutron-activated body fluids should give the most accurate data on relative neutron exposure, and measurements on blood sodium will probably give the best

results. However, calibration of body fluids for an absolute determination of exposure requires data on the incident neutron spectrum and the γ–neutron dose ratio. Area monitors spaced throughout a work area and incorporating components designed to give these data are thus indicated as an important part of an over-all personnel monitoring system.

REFERENCES

1. M. Brucer, *The Acute Radiation Syndrome, A Medical Report on the Y-12 Accident, 16 June* 1958. Oak Ridge Institute of Nuclear Studies, Inc., Oak Ridge, Tennessee, ORINS-25 (1959).
2. R. C. Rohr and H. F. Henry, *The Energy Spectrum of the Leakage Neutrons from a Homogeneous Reactor.* Carbide and Carbon Chemicals Company, K-25 Plant, Oak Ridge, Tennessee, K-736 (1951).
3. J. W. Wachter and L. C. Emerson, *Neutron Dose Calibration of Indium Personnel Dosimeters for Prompt-Critical Metal Bursts.* Y-1092 (1956).
4. F. S. Patton *et al.*, *Accidental Radiation Excursion at the Y-12 Plant, June* 16, 1958. Y-1234 (1958).
5. H. F. Henry, J. C. Bailey and R. C. Rohr, *Nucl. Sci. Engng.* **5,** No. 5 (1959).
6. H. C. Paxton, *Nucleonics* **13,** No. 10 (1955).
7. Unpublished experimental data using sucrose-urea "phantoms", radium and cobalt sources, and film badge detectors.
8. V. P. Bond, Geometrical and energy factors influencing the effect of penetrating radiation on man. *The Shorter-Term Biological Hazards of a Fallout Field* (Symposium, December, 1956). U.S. Government Printing Office, Washington, D.C. (1958).
9. S. A. Kingsbury. Unpublished preliminary work done in 1947 at the Oak Ridge Gaseous Diffusion Plant using a radium source and a G.M. tube meter.
10. J. R. Knight, Calculation of the neutron-leakage spectra and neutron dose from bare critical systems. *Transactions of the American Nuclear Society,* 1959 *Annual Meeting.* A Supplement to *Nucl. Sci. Engng.* **2,** No. 1 (1959).

Health Physics Pergamon Press 1961. Vol. 6, pp. 94–95. Printed in Northern Ireland

NOTE

Health and Safety Costs Associated with the Construction of a Nuclear Reactor

(*Received* 27 *July*, 1959)

Introduction

MANY individuals directly associated with atomic energy development have indicated the extent to which engineering criteria were influenced by the need to protect the public health and safety from the radiation hazards generated by the fission process. Persons from the diverse fields of physics, mechanical and chemical engineering, insurance, investment, government, public health and atomic energy management have repeatedly emphasized that the obtaining of protection against such atomic hazards is expensive.

Entire systems have been incorporated in nuclear reactors to provide for protection, and these systems have influenced the design of accessory or related systems. If health and safety were not the major problem in reactor design, many applications of nuclear reactors might be possible which are not seriously considered today. A nuclear reactor could consist of a few simple components: a heat source, a coolant, pipes, a turbine and associated controls. The reactor could be built almost completely out-of-doors, above ground, anywhere, at any power level and with a minimum of governmental control. This would be a power utility's executive's dream. But we must consider protection.

The basic problem

The fission process in the reactor core produces intense quantities of radiation and radioactive materials. Human beings are sensitive to any level of radiation, and while some exposure to radiation can be permitted, excessive exposure can injure and perhaps kill. The power reactor must be so engineered that each component, which affects the health hazards from routine or accidental operation, is designed to contribute toward safety. The problem is to extract useful heat and, simultaneously, minimize biological damage to human beings.

Minimizing damage to human beings influences the design of every major component in a nuclear power reactor. The entire costs of many of these can be attributed to the need to provide protection. It is not necessary to estimate the difference in costs between a special fast-closing valve in the primary coolant loop of the PWR-type reactor and a standard valve in its place. The entire coolant loop was required to isolate the steam to the turbine from the reactor system, to minimize possible contamination from radioactive materials in the coolant. Replacement of the air locks through the containment vessel with less expensive, simpler air locks does not change the fact that the entire containment vessel was required to prevent the escape of radioactive materials in the event of a reactor incident. The design effort put into air locks, the piping seals, hatch and vent covers, fast-closing exhaust ventilation ducts, special butterfly valves and special electrical connections, was required to maintain the containment vessel's integrity against leaks.

Typical of entire systems which can be "charged" against safety are primary coolant loops, containment shells, biological shields, radiation-monitoring system, the remote and large plant site and, in some instances, fuel-element cladding. There are safety factors placed on electronic control equipment, operating-temperature limits, thicknesses and nature of supporting structural materials, building foundations and auxiliary mechanical systems.

Ever present is the fear that some aspect of the safety problem may have been overlooked during design. This fear pervades the whole design problem and effects costs from the initial conception of the project to the first few months or years of operation. The consideration of health and safety looms like a ghost in every drafting room and engineering conference where atomic components or systems are designed. It forces preoccupation with extra equipment, stringent control of materials, solutions to new or unique legal questions, insurance on an unprecedented scale, and of quality and training of personnel, among many problems. Canned pumps and special valves are accepted as natural requirements, rather than as expensive alternate solutions to particular problems.

The extent to which the public accepts or rejects atomic energy power plants in a particular area is not yet evident. Only one major reactor project has been challenged on the basis of safety. Should public opposition to a reactor reach the point that no operation is possible, the *entire* cost of the construction of the plant up to that point could be considered a health and safety cost.

Obviously major systems which are directly required for radiation protection should be examined for possible reduction of costs, and several attempts have been made suggesting alternative approaches to

shielding, containment, cladding, exclusion areas around reactors, intermediate coolant loops and other significant safety systems.

Minimizing health and safety costs

The question can be asked: what are the several approaches to reactor component design which suggest themselves if health and safety costs are to be minimized? The following approaches are suggested by the above analysis.

(1) Core and fuel element design: natural uranium (possibly in the form of highly refined ore); no fuel fabrication into metal ingots or forms.

(2) Cladding: eliminate cladding by suspending the fuel in a solution which could serve as moderator, neutron reflector and perhaps the coolant as well (possibly heavy water).

(3) Intermediate coolant loops: eliminate intermediate coolant loops; use coolants which minimize neutron activation.

(4) Fission product control: continuous removal of fission products.

(5) Shielding and containment: combine the containment with the shielding.

(6) Standardization: fix design criteria for all components, systems or auxiliaries to test against contribution to safety; minimize use of special components.

The elimination of the intermediate coolant loop has been accomplished in the boiling-water reactor system, which uses highly purified water as the coolant —which is actually boiled within the reactor core and the steam passed directly to a turbine. Other approaches to the above suggestions have been attempted on an individual basis. The essential ingredient in all of these approaches is to optimize health and safety costs, particularly identifying those which cause escalating increases in other component design costs. That reactors containing fundamental design approaches similar to those developed above are in the process of being planned or built would seem to indicate that over-all costs may be significantly reduced, and that economical, competitive, nuclear power is not impossible, through optimization of identified health and safety costs.

Conclusions

(1) Health and safety requirements underly the design, or testing, of almost every nuclear power reactor component or system design decision.

(2) Health and safety requirements underlie almost every aspect of government regulation of atomic energy, including the creation of a unique federal government supervision.

(3) Health and safety requirements underlie many aspects of operating costs of a nuclear reactor, including specialized personnel, extraordinary insurance, and special labor and public relations programs.

(4) The composite cost of each of the individual costs which are attributable to health and safety may account for from 20 to almost 100 per cent of the entire nuclear power plant cost.

(5) The composite cost of the specific health and safety requisites for a pressurized water-type reactor is estimated to be a *mininum* of 39 per cent of the reactor portion of the construction costs, using data available at the time this summary was prepared on costs of systems and components.

(6) Hidden safety costs, when added to the specific ones, may result in a *minimum* of 65 per cent of the construction costs of a pressurized-water reactor. For the boiling-water reactor, the health and safety costs add up to 57 per cent of the total construction costs.

(7) There are approaches to reactor design which suggest themselves if safety costs are to be reduced—which should result in a reduction of the cost of electricity produced as a result of nuclear fission.

(8) There are reactor systems which are being considered and developed which are applying these approaches. Some have progressed to the point where competitive nuclear power is predicted from plants now under construction.

(9) If all approaches suggested herein could be combined, in perhaps a new reactor type, nuclear-produced electricity should be obtainable at lowered costs—without jeopardizing necessary safety requirements.

(10) Recognizing the effect on the design and costs of subsequent components from increasing a safety factor in an initial, determinant, system should warn the design engineer to make such initial-design decisions only after complete evaluation of all possible effects.

(11) Standardization of reactor designs, at least in terms of safety criteria upon which to make decisions, is suggested.

SAUL J. HARRIS

Atomic Accessories, Inc.
Valley Stream, New York

HEALTH PHYSICS SOCIETY
PLACEMENT COLUMN

Orders for ads to be sent prepaid to the Placement Committee at the address below. Ads will appear in the next issue following receipt.

1-Time Charges:

Position wanted: $0.25 per word ($4.00 minimum)
Position offered: $1.00 per word ($15.00 minimum)

Replies to employer or to
Mr. Joseph A. Lenhard,
Chairman Placement Committee H.P. Society,
Research and Development Divison, USAEC
P.O.B. E, Oak Ridge, Tenn.

POSITIONS OFFERED

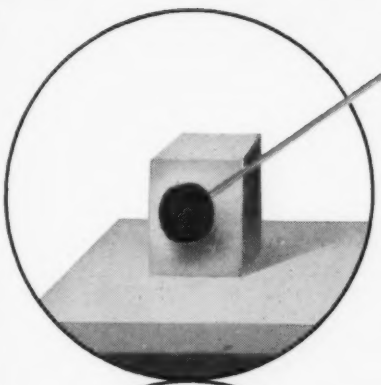
fee paid positions for
HEALTH PHYSICISTS
Write for Application • No Resume Required • Confidential Handling
We are a national placement service with technical experience, dedicated to people in the nuclear field . . . wherever radioactivity is a factor.
ATOMIC PERSONNEL, INC.
Suite 1207-R, 1518 Walnut Street, Philadelphia 2, Pa.
An Employment Agency for the Nuclear Field

AT LAST! . . . a specific tool for wipe testing of sealed radioactive sources

Wipe-Swipe
SAFETY WAND

Source is our Product

Another RDCo RADIATION-DETECTION-COMPANY First!



SAFE • EFFICIENT
• EASY TO USE •

Telescopic handle extends to 42"

RADIATION DETECTION COMPANY PROVIDES RADIOMETRIC ANALYSES!

ALL SAMPLES WILL BE ANALYSED AND MAILED BACK TO YOU WITHIN 24 HOURS

"Wipe-Swipe" is specially designed to meet the periodic wipe test requirements of the AEC Isotopes Division. A remarkably handy tool for leak tests of sealed sources containing beta-gamma and alpha-emitting radioisotopes, it offers advantages in added safety, convenience and efficiency. In addition, "Wipe-Swipe" is of simple and sturdy construction, contains no moving parts and should last indefinitely without maintenance or service.

Filter paper can be inserted or removed quickly and easily: the wipe retains its original shape — a great advantage in ease of counting.

Write for our bulletin on "How to make a leak test with Wipe-Swipe."

Radiation Detection Co. offers a COMPLETE HEALTH PHYSICS SERVICE

- BIOASSAYS
- FILM BADGE DOSIMETRY
- X-RAY CALIBRATIONS
- RADIATION SURVEYS
- HEALTH PHYSICS CONSULTATION
- ENVIRONMENTAL ANALYSES

Inquiries are invited on our latest development The R-D CASUALTY BELT

The R.D belt is an improved means of determining neutron exposure from criticality accidents. Providing accurate attenuation data as well as traditional measurements, the belt detects the neutron dose from any angle. Neutron exposures as low as 10 rad. can be detected, while the upper limit extends far beyond the lethal range. Belts are of leather and similar in appearance and serviceability to good quality dress belts. Sizes range from 30 to 46 in black or brown.

Radiation Detection Co.

4047 Transport St., Palo Alto, Calif. • Phone DA 3-1279

Daylight on Day Hospitals
—Recent Reviews of

The Day Hospital Movement in Great Britain

JAMES FARNDALE

"This interesting book falls into two parts. The first part traces the development, organisation and functions of day hospitals and day centres—both psychiatric and geriatric—with particular emphasis on administrative, economic and social aspects, while the second part gives a detailed account of 65 day hospital and day centres which the author visited in 1958–59. During that year Farndale was seconded from his position as deputy secretary to the Board of Governors of the Bethlem Royal Hospital and the Maudsley Hospital, London, to carry out a research project at the University of Manchester. The book is, in fact, the report of that project, which was carried out with the "agreement, interest and support of the Ministry of Health and certain professional, voluntary and charitable organisations.

"Subjects suggested for study included the cost of day treatment in relation to that of inpatient treatment, the question of whether any saving might result to the National Health Service by treating more psychiatric patients by the day, the effect of day hospital provision on the demand for inpatient beds, and whether day hospitals are relieving or would relieve the overcrowding in some mental hospitals.

"Farndale makes no exaggerated claims for the success of day hospitals, rather is there a note of caution and reservation. But though he admits that the movement has so far developed on a very small scale, he points out that the work carried out by day hospitals and day centres "is impressive". "There appears to be a need for more . . . . . and they deserve to be widely developed." Points suggested in favour of this development are: many patients would prefer treatment as day patients to treatment as inpatients if offered the choice: many doctors think that day hospitals are a satisfactory substitute for inpatient treatment, and some consider that it is a better method for certain patients; again many doctors consider day hospitals to have medical and therapeutic advantages; and the public themselves are showing an increasing desire for day and evening psychiatric treatment.

"Psychiatric day hospitals, says Farndale, could make their maximum contribution when in close touch with a psychiatric hospital or psychiatric inpatient unit, though not necessarily 'attached' thereto.

"The book carries a preface from the Hon. W. S. MacLay, senior medical commissioner of the Board of Control, who commends it to all who are planning day hospitals as part of their service to the community."

Hospital and Health Management, June, 1961.

". . . . . all those who are concerned with the operation or planning of day hospitals will find this an invaluable work of reference."

American Journal of Psychiatry

430 pp. 84s. net ($15.00)

PERGAMON PRESS

Radiation Monitors

FOR AIR, WATER, GAS

Units or systems to solve practically any monitoring problem. Model AM-33R is the first air monitor to solve the natural radon progeny problem by providing direct identification of long half-life emitters.

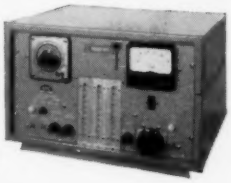
AM-33R $6,580.00
Others from $1,445.00

γ ALARM SYSTEM

This maintenance-free "Gammaguard" provides an alarm for any area where fissionable materials are stored, used or processed. Eliminates central power supply. Works alone or as part of a system. GA-2 $595.00

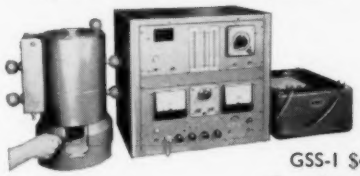
Proportional Counting Systems

PC-3A $1,395.00



The only instrument that counts every alpha and beta emitted in a 2 pi angle from prepared samples. Ideal for C-14, Ca-45, P-32 and H-3. No window absorption. A four pi model also is available.

γ Spectrometer System



GSS-1 $4,180.00
(Complete System)

The one system that eliminates "dark current" defect. Combines better resolution with greater precision over wider range than any other system—at any price.
Range: 7 kev. to 5 mev.

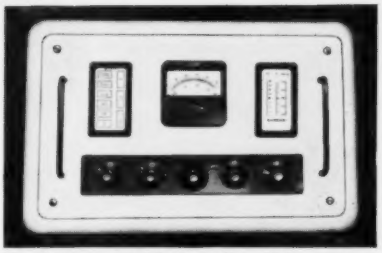
All prices are F.O.B. Indianapolis

WRITE FOR NMC FREE LITERATURE

NUCLEAR MEASUREMENTS CORP.
INDIANAPOLIS

2460 N. Arlington • Indianapolis 18, Indiana
International Office 13 E. 40th St., New York 16, N.Y.

Highly accurate dose and dose-rate measurement—



OVER AN EXTREMELY WIDE RANGE

The Baldwin IONEX Ionisation Meter measures DOSE from 300μr—1000r. and DOSE RATE from 30μr/min.—1000r/min.

The Baldwin IONEX Ionisation Meter is a precision D.C. amplifier for measuring dose and dose-rate of ionising radiations over a very wide range of energy and intensity. A small thimble chamber of approximately 0.6 cc. is attached to the end of a flexible cable to measure dose distribution, depth dose, half value thickness, and to calibrate other instruments.

For measuring soft X-rays, alpha and beta radiation, radiographic exposures, and for checking contamination, a nominal 600 cc. chamber with a window of 0.005 in. Styrafoil (equivalent to approximately 1 cm. of air) is used. Facilities are provided for the fitting of a recorder where required.

Write now for Leaflet HP 37/118 which gives complete, easy-to-read information.

INSTRUMENT DIVISION

Baldwin Industrial Controls

Baldwin Instrument Company Ltd., Dartford, Kent

Telephone: Dartford 26411 & 20948 Cables: Baldwin, Dartford

Telex: Baldwin Dartfd. 25236 *A HARPER GROUP COMPANY* B37

featuring ultra-high stability and precision switching

E.H.T. SUPPLIES NE 5302 & NE 5303

•VOLTAGE RANGES

NE 5302: 500–1500 volts D.C.: max. output current 5 mA.
NE 5303: 1000–5000 volts D.C.: max. output current 3 mA.

•SPECIAL PRECISION SWITCHING

Both units feature use of switched precision resistors for coarse steps of voltage adjustment, and 0.1% ten-turn potentiometers for fine voltage control, thus ensuring accurate resetting of the output voltages to 0.1 volts.

•STABILITY AGAINST FULL LOAD

For both units, 50 parts per million measured at full output voltage.

•STABILITY AGAINST LINE VARIATIONS OF ±10%

For both units, ±50 parts per million measured at full output voltage.

•NOISE AND RIPPLE LEVELS

Less than 5 millivolts for any voltage setting.

Also available, the following companion units:

Non-overloading Linear Pulse Amplifier NE 5202; Preamplifier and cathode follower NE 5202A; Single-channel pulse height-selector NE 5102; Scanning Scintillation Spectrometer NE 8601; Ratemeter NE 5401

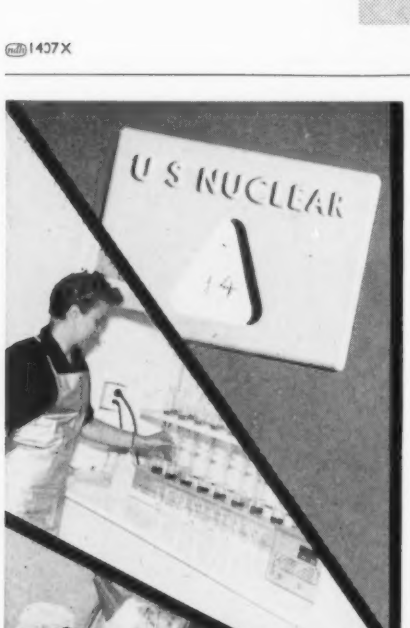
Write for new catalogue, Price List

Nuclear Enterprises (G.B.) Ltd

Bankhead Medway, Sighthill, Edinburgh 11, Scotland. (Tel. CRA 5262)

Associate Company: Nuclear Enterprises Ltd.
550 Berry Street, Winnipeg 21, Canada

1407X



ONE RELIABLE SOURCE

for all

RADSAFE

SERVICES & EQUIPMENT

including

Health Physics Consulting
Low Level Counting
Leak Testing of Sources
Bioassay
Plant Survey
Decontamination
Film Badge
Training Courses
Environmental Surveys
Effluent Analyses
Alpha, Beta, Gamma & Neutron Monitors
Air Samplers
Alpha, Beta, Gamma & Neutron Calibration Sources

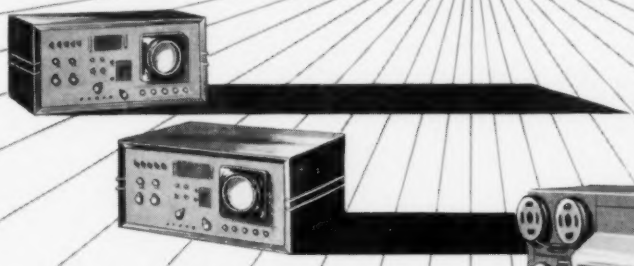
write for catalogs and quotations

Industrial and Research Radioactive Sources, Tracers and Associated Equipment are also offered.

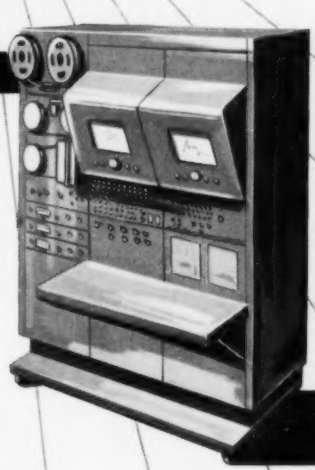
U.S. NUCLEAR CORPORATION

Box 208 Burbank, Calif. Vi 9-6176

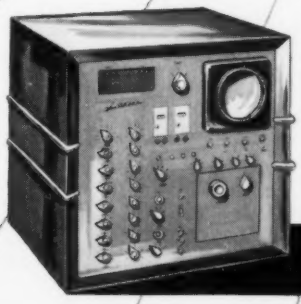
VICTOREEN *First with the Finest in Analyzers*



Models ST-200, ST-200D, ST-400, ST-400D



Multidimensional Series



"M" Series — 400 and 800 Channels (1 to 8 inputs)

Victoreen's fine family of analyzers is widely recognized as being unparalleled in . . . versatility . . . long term usefulness . . . performance . . . value.

Their superior design concepts provide the highest degree of linearity and freedom from data shift, independent of counting rates. Linear selection memory circuitry assures rapid and reliable data handling. Multiple input configurations meet any user requirement.

These Victoreen analyzers provide all of the features most desired — and are the *only* analyzers which bear the name Tullamore — the Hallmark of Quality.

A-5182A

Service by
RCA SERVICE COMPANY
A Division of Radio Corporation of America

Victoreen

5806 HOUGH AVENUE • CLEVELAND 3, OHIO
EXPORT: 240 WEST 17th ST. • NEW YORK 11, NEW YORK

VICTOREEN—WORLD'S FIRST NUCLEAR COMPANY

PERGAMON PRESS

specialized journals for *specialized advertising!*

— — — — — — — — — — — —

Advertise in HEALTH PHYSICS

Space is available to all manufacturers of survey instruments (geiger counters, ion chambers, scintillation counters, proportional counters, fast neutron survey instruments, alpha monitors, etc.); personnel monitoring equipment (film badges, pocket ion chambers, pocket alarm meters, film badge service, personnel monitoring film, etc.); area monitoring equipment (constant air monitors, water monitors, fallout-trays, etc.); building monitors (alarm meters, filter paper, scalers, air samplers, instruments for assessing levels of α, β, γ or n activity, hand and foot counters, clothing monitors, laundry monitors, etc.); counting room equipment (scintillation counters, ion chambers, proportional counters, 4π counters, gas flow counters, coincidence counters, range analyzers, multiple channel analyzers, etc.); and a vast amount of other equipment such as total body counting equipment, stack filters, protective clothing, shielding equipment, X-ray equipment, dark-room photographic equipment and remote control equipment, protective clothing, shoe covers, etc., etc.

THIS IS ONE OF MORE THAN 70 INTERNATIONAL SCIENTIFIC JOURNALS. SPECIAL REDUCED "COMBINED RATES" ARE OFFERED TO MANUFACTURERS ADVERTISING IN MORE THAN ONE JOURNAL.

— — — — — — — — — — — —

For details WRITE OR PHONE to

R. D. MILLER
Advertisement Manager
PERGAMON PRESS

4 Fitzroy Square,
London W.1
Phone: EUSton 4455

122 East 55th Street,
New York 22, N.Y.
Phone: Plaza 3-9651

— — — — — — — — — — — —

TRY COLOUR FOR ADDED PRESTIGE

AT YOUR FINGERTIPS — A PERSONAL WARNING SYSTEM

MODEL L-135
PERSONAL RADIATION MONITOR

For those who work with potentially hazardous radiation.

The perfect companion instrument to your Landsverk Dosimeter. It provides an audible and visual alarm when exposed to above normal radiation intensity. A sensitive subminiature geiger counter complete with transistor power supply, transistor amplifier and alarm circuit that do not block out at high intensity.

- Indicates backgrounds to 10 r/hr
- Model L-135A up to 100 r/hr
- Long battery life
- 5½" long by ¾" diameter
- Weighs only 3 ounces

Landsverk
ELECTROMETER COMPANY

641 SONORA AVE. • GLENDALE 1, CALIFORNIA

PLENUM PRESS

BIOLOGICAL EFFECTS OF MICROWAVE RADIATION

Volume I

Proceedings of Fourth Annual Tri-Service Microwave Conference, held August 16—18, 1960, New York University Medical Center.

Sponsored by Rome Air Development Center, Air Research and Development Command, United States Air Force, Griffiss Air Force Base, New York.

Edited by Mary Fouse Peyton, Department of Industrial Medicine, New York University Post-Graduate Medical School.

The papers in the present volume range from discussions of fundamental considerations of the molecular responses of living tissues to microwaves, through reports concerned with the physical characteristics and measurement of microwave energy, to the organizational and administrative problems of protection. This volume will be of considerable value to all those concerned with radar hazards and their prevention.

327 pages Complete contents upon request **$10.00**

PLENUM PRESS
227 W. 17th St. • New York 11, N. Y.

ENVIRONMENTAL HEALTH ENGINEER

DOW

Opening in environmental research section of corporate Biochemical Research Laboratory.

Position involves primary responsibility for measurement, evaluation, and control of plant environmental conditions in the field of radiation, but also includes investigation of such other factors as exposure to chemicals, heat, light, and noise. Requires undergraduate training in chemistry or chemical engineering, and M.S. training, or experience, in health physics or industrial hygiene.

Salary commensurate with training and experience. Excellent educational, recreational, and living facilities at Midland, Michigan. Relocation expenses paid. Send resume to:

James E. Campbell
P.O. Box 468
The Dow Chemical Company
Midland, Michigan

PERSONAL RADIATION MONITOR

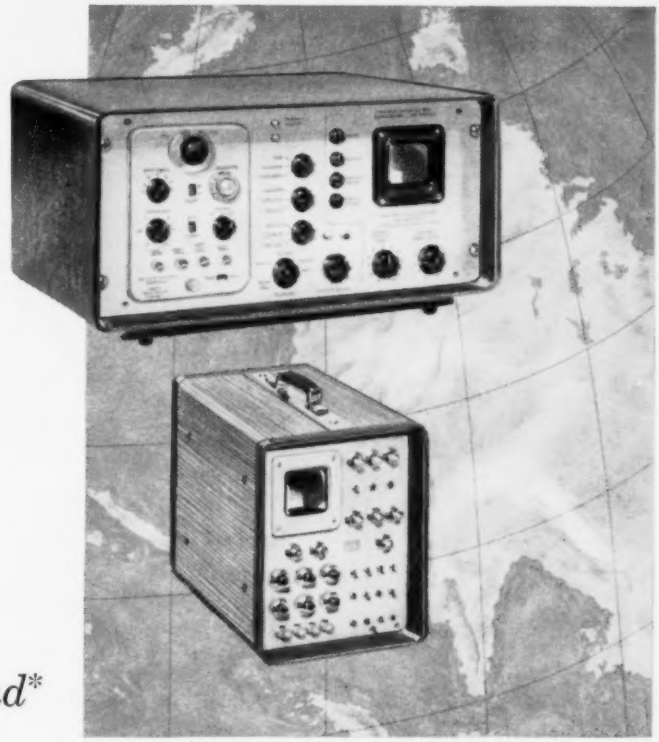
- Loud chirping signals
- Flashing neon lamp
- Unique "on-off" device
- Lightweight . . . only 3½ oz.
- Long battery life
- Based on O.R.N.L. design

Model PRM-253 "Chirpee" is a miniature, lightweight (3½ oz.) radiation monitor that warns the user when he encounters an unexpected radiation field. It features visible and audible warning signals . . . a flashing neon lamp and a "chirping" sub-miniature speaker. Both are activated simultaneously, at a rate proportional to radiation intensity.

Write for Bulletin H53

ATOMIC
accessories inc.
Subsidiary of Baird-Atomic, Inc.
817 W. Merrick Rd., Valley Stream, N. Y.
Represented Nationally by
Baird-Atomic Sales Offices

*What's good for nuclides in New Haven is good for those in New Zealand**



We think the reason people all over the world buy TMC pulse analyzers is pretty much the same reason they're widely purchased and used here: competently designed, bug-free circuits . . . straightforward operation with the same performance and stability today as yesterday . . . easy access to sub-assemblies and uncomplicated servicing if needed. Another way of putting it is the instruments give the user the information he wants in *his* work — with predictable behavior—regardless of where his site, lab or plant may be located. Here are two current examples:

The TMC CN-110 256 channel analyzer offers 7 interchangeable plug-in logics, including pulse height, time of flight, pulsed neutron, multiscaler, mass spectrometer, and coincidence pair. This widely used and thoroughly proven analyzer (over 100 units have been delivered) employs all-transistorized circuitry. Analog, binary, octal and decimal readout may be used. Data can be recorded on strip chart or X-Y recorders, printed paper tape, punched paper tape, or punched cards.

The Model 404 is a compact, 400-channel analyzer you can use anywhere there's a wall outlet and one square foot to put it down. It has a magnetic core memory that can be used in sub groups of two or four; four separate inputs and associated amplifiers; internal pulse routing circuity; pushbutton data transfer and display overlap; power requirement of *only 25 watts*, and many "system" advantages. While its versatility is a little less than the CN-110's, so are its size, price and purpose quite different from the 110's. Each does its *own* job well.

**Also wherever TMC Pulse Analyzers are used . . . in Canada, Brazil, Australia, Japan, Yugoslavia, France, Italy, Germany, Belgium, Sweden, Denmark, Switzerland, Israel, Formosa . . . as well as the United States.*

WRITE FOR LITERATURE.

TMC **TECHNICAL MEASUREMENT CORPORATION**
441 WASHINGTON AVE., NORTH HAVEN, CONN. • CE 9-2501

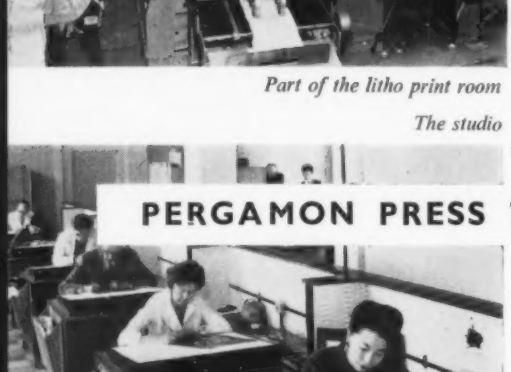
A SKILLED TECHNICAL WRITING SERVICE FOR SCIENCE AND INDUSTRY

The technical editing department

One section of the drawing office

Part of the litho print room

The studio



Do you need a really fast, efficient, 'one source' technical writing, illustrating and printing service? The Pergamon Press Technical Writing Division offers a fully integrated service, capable of undertaking the complete design and production, including translation from and into foreign languages, of all types of technical printed matter, and especially user/operator handbooks, instruction and maintenance manuals, sales brochures, catalogues and company reports.

The Division is staffed by experienced, highly qualified technical writers, draughtsmen and typographic designers, and in addition can consult the several hundred internationally eminent authors and editors who publish with the Press, and obtain their advice and services in every specialized sphere of science, medicine and technology.

We offer keenly competitive prices, high quality and speedy delivery, and shall be pleased to submit quotations for all or any one of our services. We guarantee complete security when handling work of a confidential nature.

For immediate attention to your requirements please write or telephone

JOHN BURLS, B.Sc., *General Manager*

PERGAMON PRESS TECHNICAL WRITING DIVISION

4/5 FITZROY SQUARE,
LONDON W.1
Telephone: EUSton 4455

Editorial and sales offices also at:

Headington Hill Hall, Oxford
122 East 55th Street, New York 22, N.Y., U.S.A.
24 Rue Des Ecoles, Paris V^e^ Kaiserstrasse 75, Frankfurt am Main

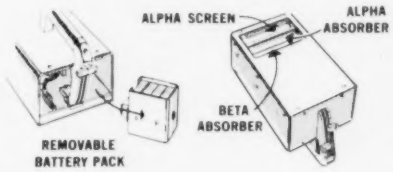
NEW MODEL SURVEY METERS

...with important new features for Health Physicists!

JUNO
measures and distinguishes between alpha, beta, and gamma radiation

New Model 7 JUNO
is DEPENDABLE even under high humidity conditions

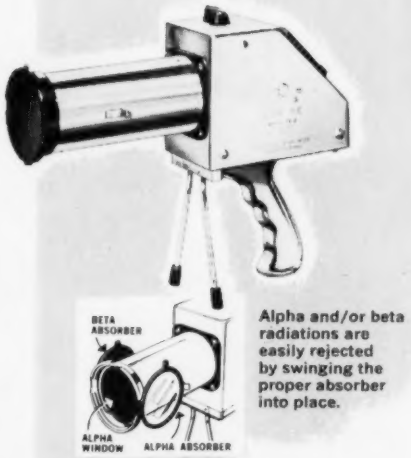
The Model 7 Juno is T/A's new improved version of the original Hanford instrument. It features a hermetically sealed switch box and desiccant cartridge, thus insuring high efficiency performance under adverse humidity conditions. Additional features include a high impedance circuit, a large open-face meter, and a new 800-hour battery pack.



New Model CP-3 CUTIE PIE
has built-in alpha and beta absorption filters!

This portable survey meter has 3 full-scale ranges of 50, 500, and 5000 mr/hr and is especially useful in detecting alpha, beta, or gamma radiation in such inaccessible spots as corners, behind pipes, etc. It incorporates new circuitry, plus a battery pack with 800 hours of operating life.

Write for Bulletin No. 180 (Juno) or Bulletin No. 165 (Cutie Pie)



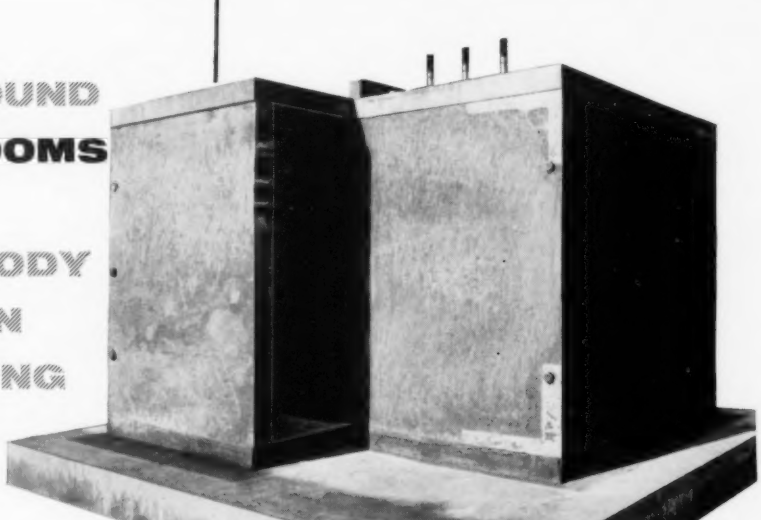
Alpha and/or beta radiations are easily rejected by swinging the proper absorber into place.

TA **TECHNICAL ASSOCIATES**

Instrumentation for Nuclear Research

140 WEST PROVIDENCIA AVENUE—BURBANK, CALIFORNIA

LOW
BACKGROUND
STEEL ROOMS
FOR
HUMAN BODY
RADIATION
MONITORING



as already supplied to Windscale, Karlsruhe, Adelaide and Dounreay

SAUNDERS - ROE & NUCLEAR ENTERPRISES LTD.
NORTH HYDE ROAD, HAYES, MIDDLESEX. TELEPHONE: HAYES 3800

Health Physics Pergamon Press 1961. Vol. 6, pp. 97–102. Printed in Northern Ireland

SOME FUNDAMENTAL ASPECTS OF MARINE ECOLOGY IN RELATION TO RADIOACTIVE WASTES*

MILNER B. SCHAEFER
Inter-American Tropical Tuna Commission, Scripps Institution of Oceanography, LaJolla, California

(*Received* 10 *October* 1960)

THE MARINE ENVIRONMENT AND ITS SUBDIVISIONS

THE oceans of the world cover about 71 per cent of the earth's surface to an average depth of 3800 m. The ocean basins are characteristically bordered by a continental shelf which slopes out gently to a depth of about 200 m. Beyond the shelf, the continental terrace slopes much more abruptly to the deep sea floor at depths of 4000 m or more. The average width of the shelf is about 30 miles, varying from practically nothing off mountainous coasts to several hundred miles in such places as the China Sea. The shelf is not always smooth, but it is often intersected by submarine valleys and canyons. In the deep ocean there are high mountains and deep trenches, larger than any on land. Some of the mountains rise to the surface, forming islands, or to near the surface, forming "banks" and "shoals".

The waters of the ocean form, essentially, a two-layer system. The relatively thin upper layer varies from a few meters to about 200 m in thickness, averaging about 75 m. In this layer, vertical mixing caused by winds is relatively rapid and complete, so that temperature, salinity, and density are nearly uniform from top to bottom. In it are found the fast, wind-driven currents which are the "surface" currents shown on many charts. Below the mixed layer is a zone, extending down to 500 m in some places, within which the temperature decreases and density increases rapidly with depth. This thermocline, or pycnocline, separates the upper mixed layer from the layer of intermediate and deep water extending to the bottom, where the vertical gradients of decreasing temperature and increasing salinity and density are very gentle. Both vertical and horizontal movements in the deep layer are much slower, in general, than in the upper layer. The strong density gradient across the thermocline inhibits physical transport across it, thus there is a partial barrier between the upper mixed layer and the lower layers. There is, however, some interchange of water and its dissolved nutrient salts across the thermocline. Indeed, if there were not, the upper layer could not continue to support much life.

The energy that drives the whole life cycle in the sea, as on the land, is sunlight. This energy is bound chemically in organic compounds by the photosynthesis of plants, and is passed along through the food web in the food of all higher organisms. The zone of the sea to which sufficient light penetrates to support photosynthesis is only about 100 m deep, even in very clear waters, and is often less. Thus most of the photosynthesis takes place in the upper mixed layer, and mostly by microscopic floating plants,

* Presented at the Symposium on Waste Disposal, Fifth Annual Meeting of the Health Physics Society, Boston, Massachusetts, June, 1960 Contribution from the Scripps Institution of Oceanography, New Series.

the phytoplankton. Only along the shallow margins of the sea are there large algae, and other plants analogous in size to plants on land. In order to grow, plants require, in addition to sunlight and water (of which there is of course plenty in the sea), certain chemical nutrients such as phosphates, nitrates, etc. Now, due to gravity effects on the dead plants and animals and their excreta, there is a continual rain of organic material from the upper layer to the deeper layers of the sea. Some of this material arrives at the very bottom of the ocean, even in its deepest parts, since living organisms, which depend ultimately on the plants near the surface, are found at all depths. This fallout of organic matter continually removes the plant nutrients, which are, by bacterial action, regenerated as inorganic compounds in the deep water, and must be returned to the sunlit zone to fertilize it.

Although there is probably some upward mixing of the water across the thermocline almost everywhere, the restoration of nutrient-rich water from below is most intense where the sea is "plowed" by certain physical processes. In some places winds drive the surface water away from the coast or an internal boundary and nutrient-rich waters well up from mid depths. At high latitudes, surface waters in winter are cooled to near freezing, sink, and mix with deeper waters. At boundaries of ocean currents, violent mixing brings up deeper waters. Over the continental shelf, the materials from the upper layer fall on to the bottom not far below, and are thus readily accessible to upward mixing. Such regions are the green pastures of the sea, supporting abundant life and major sea fisheries. Other regions, where the surface waters have been depleted of nutrients are biological deserts, supporting only very sparse populations.

Man's food harvest from the sea, which is the important route by which radioactive nutrients are returned to him, is taken almost entirely from the upper mixed layer of the open sea or from inshore and coastal waters out to the edge of the continental shelf, although some fisheries extend to depths of 500 fathoms, and some of the harvested fish populations perform migrations to deeper waters.

From the viewpoint of the physical and biological processes involved in returning to man radioactive materials introduced into the sea, there are several important sub-divisions of the marine environment.

(1) *Near-shore areas*, including the close inshore zone of the open coast and areas which are partially enclosed by land, i.e. harbors, bays, estuaries and lagoons. In these areas dispersion of wastes by physical processes is often restricted, and, since they are shallow, the material that falls out on the bottom does not become inaccessible to the organisms. In this environment occur the large sedentary populations of oysters, clams, and seaweeds used by man, and it is an essential nursery ground for shrimp, menhaden and many other offshore commercial species, as well as the habitat of many fishes taken for food by man.

(2) *Continental shelf and the overlying waters.* Removal of waste materials by diffusion and current transport is generally greater here than in near-shore areas, but less than in the open sea off-shore. Sedimentation to the bottom may not remove elements from the biosphere in this region, since food organisms taken by man live all the way to the bottom. On the bottom are scallops, shrimp, crabs, and lobsters and also populations of both moving and sedentary invertebrates which are food for important commercial fish populations such as flounders, cod and haddock which live on or near the bottom. Some of the commerical species are fairly sedentary, but some perform extensive seasonal migrations to and from deeper waters. Some commercial species are found out along the continental terrace to depths as great as 1000 fathoms, or more.

(3) *The deep sea beyond the continental shelf.* Radioisotopes return to man from this region through the seafood harvest which depends almost entirely on organisms inhabiting the upper mixed layer or the region of the thermocline. Thus isotopes which are introduced into the deep water, or which are removed to the deep water from the surface, are mostly removed from the domain of man's food. The mean time for interchange between deep water and the surface layer is long, being of the order of at least several decades and probably several

hundred years, so that there is much time for dispersion and decay. Elements which sediment out on the bottom are removed for even longer periods, perhaps permanently. Dispersion in the upper layer is also rapid, leading to quick dilution. However, there is some additional interchange between the deep and surface layers due to migration of organisms, and this can be in some circumstances more important than the physical exchange.

The food web in the sea

One of the important differences between life in the sea and on land is the much greater length of most food chains in the former. On land the chains are mostly short, such as grass to cows to men. In the sea, particularly in the pelagic realm, food chains are much longer. The plants are mostly microscopic phytoplankton, which is eaten by tiny herbivorous animals which are eaten by small carnivores that are consumed by larger ones. Although man consumes some organisms low in the food chain, most of our seafish harvests is from animals two or more steps above the primary plant producers. At the same time, however, some of the animals take up minerals, both dissolved and particulate, directly from the sea water, so that radioisotopes can return to man by a shorter route than that starting with the plants. These complexities, and our lack of knowledge concerning them, are among the principal handicaps in precise evaluation of methods of sea disposal of radioactive wastes.

More detailed discussion of the foregoing matters, which I have reviewed very briefly, can be found in REVELLE *et al.*[1], BROWN *et al.*[2], CARRITT *et al.*[3], PRITCHARD *et al.*[4], REVELLE and SCHAEFER[5], SCHAEFER[6], KETCHUM and BOWEN[7].

With this general background, I would now like to discuss in a little more detail some of the processes in the ecosystem which are of importance in relation to radioactive wastes.

Uptake and retention of radioisotopes by organisms used as food for man, and the evaluation of permissible concentrations in the environment

Radioactive isotopes in the sea affect man primarily through his ingestion of marine organisms in his food. Thus, quantities of radioactive materials that can be safely introduced into the sea depend on the amounts that come to lodge in such marine organisms, and the permissible rate of intake by man. The last can be determined from the permissible rates of ingestion and body burdens of various radioisotopes published by the national and international committees on radiation protection. In making estimates of permissible concentrations in sea water, account must be taken of the facts that sea water is a solution of almost all the elements; that concentrations of many elements are known and that some of them are very nearly in constant proportions; and that marine animals concentrate, often by very large factors, some isotope both radioactive and non-radioactive. It should also be noted that organisms do not significantly distinguish between the radioactive and non-radioactive isotopes of a particular element, so long, at least, as the radioactive and non-radioactive species are in the same chemical form.

Various elements, in addition to the major organic constituents carbon, hydrogen, and oxygen, are essential to life processes, such as nitrogen, phosphorus, calcium, strontium, copper, zinc, cobalt and iron. These may be obtained by organisms above the plants in the food chain either from their food or by direct uptake from the water. Since the chemical requirements and concentrating ability differ for different kinds of organisms, the fluxes of various elements differ from species to species and at different parts of the food web. The concentrations of various elements in sea water and in different kinds of organisms, to the extent they are known, have been tabulated by REVELLE *et al.*[1], PRITCHARD *et al.*[4] and REVELLE *et al.*[8]. Some elements which are abundant in sea water, for example sodium, occur in organisms at lower concentrations than in the water; they are selected against. On the other hand, elements such as phosphorus, that are essential to the organisms, but are scarce in the sea, are concentrated by several orders of magnitude.

The length of time an organism retains the average atom of a given element varies greatly from one element to another and with different

organisms; this is often given as the "biological half-life". Much is known about the retention times of various elements in man[9], but there are few data for marine organisms.

Hazards to humans from radionuclides result from radiation to the gastro-intestinal tract as the food passes through it, and from radiation to other organs in which they accumulate. As indicated in NBS Handbook 69[9], the limiting factors for many isotopes, such as Cr^{51}, Fe^{59}, Co^{60}, Zr^{95}, Nb^{95}, Ru^{106}, Ce^{144} and Ta^{182}, is the dose to the gastrointestinal tract; for these we need to limit the concentration per unit volume in the seafood. For others, such as P^{32}, S^{35}, Fe^{55}, Zn^{65}, Sr^{90}, I^{131} and Cs^{137} the body burden is limiting; for these the specific activity of the seafood is of primary interest.

We may formulate some of these matters quantitatively following REVELLE *et al.*[8]

Designating by I_r and I_n the concentrations of radioactive and non-radioactive isotopes, it may be shown, that at equilibrium, a man getting all of the elements from seafood, will have a specific activity related to the specific activity in the seafood by

$$\frac{I_{rb}}{I_{nb}} = \frac{I_{rf}}{I_{nf}}\left(\frac{B}{K+B}\right) \tag{1}$$

where b denotes the human and f the seafood, while B is the rate constant for biological elimination of the element in man and K is the radioactive decay constant.

Similarly, when the marine food organism accumulates an element directly from seawater, in equilibrium,

$$\frac{I_{rf}}{I_{nf}} = \frac{I_{re}}{I_{ne}}\left(\frac{B_f}{K+B_f}\right) \tag{2}$$

where e denotes the sea water environment and B_f is the rate constant for biological elimination of the element in the fish. Then,

$$\frac{I_{rb}}{I_{nb}} = \frac{I_{re}}{I_{ne}}\left(\frac{BB_f}{K^2+KB_f+KB+BB_f}\right) \tag{3}$$

For elements which reach man following several steps in the food chain an analogous expression with more terms within the parenthesis can be written, but we know yet so little about the biological half-lives and mode of accumulation in marine organisms of most elements that this is only an exercise without much useful application.

In the absence of knowledge, we make the conservative assumption that B_f is large with respect to B or K, or that the hold-up time in the marine organisms is short. Thus, for elements of long radioactive half life, where $K \ll B < B_f$, approaches

$$\frac{I_{re}}{I_{ne}} \doteq \frac{I_{rb}}{I_{nb}} \tag{4}$$

so that the permissible specific activity in the ocean is about equal to that in the critical human body organ for isotopes of long radioactive half-life. Where $B_f \gg K \gg B$, we obtain from equation (3)

$$\frac{I_{re}}{I_{ne}} \doteq \frac{I_{rb}}{I_{nb}}\left(1+\frac{K}{B}\right) \tag{5}$$

Thus, for isotopes of short radioactive half-life and long biological half-life in the human body, the permissible specific activity in the ocean is much greater than in the body. If the biological half-life in the marine food organisms is also known, the estimate of permissible specific activity in sea water can be further increased.

Where radiation to the gastrointestinal tract is critical, we are concerned with the concentration per unit volume of seafood or I_{rf}. It may be seen from equation (2) that taking I_{nf}/I_{ne} as the concentration factor (F) from sea water to seafood, we have

$$I_{rf} = F \,.\, I_{re}\left(\frac{B_f}{K+B_f}\right) \tag{6}$$

I_{rf} is comparable to the maximum permissible value for drinking water for the gastrointestinal tract given in Handbook 69. Given an estimate of seafood eaten, per man per day, and taking the assumption of 15 l. of drinking water a week on which the MPC's for water were based, one may then calculate I_{re}, the permissible activity in the seawater.

REVELLE *et al.*[8] have tabulated for a number of isotopes the specific activity in sea water, and concentration factors in marine organisms, and the resulting permissible concentrations in sea water, both for cases where the gastrointestinal tract is critical and where the body burden for some organ is critical.

It should be noted that the concentration factors tabulated in this and other references earlier cited have usually been the highest observed in seafood; furthermore, in some cases they have been based on the observed concentrations of radioisotopes in sea water and organisms rather than on the non-radioactive species, and thus (as may be seen from equation (2)) may be overestimated. This is, again, a conservative approach made necessary by our lack of knowledge of the marine ecological systems.

Effects of organisms on the distribution of elements in the sea

The foregoing type of approach can provide estimates of permissible concentrations of radioisotopes in the sea water environment of the marine food organisms, but to relate this to waste disposal practices we must also know about the transport and dispersion of the material put in the sea. This is not a function only of physical parameters, but can also be significantly influenced by the marine biosphere. Those elements of which a large proportion is cycled through organisms can have their spatial distributions profoundly modified by the biosphere, so that they are quite differently distributed in the sea than those elements whose distributions are determined only by physical and inorganic chemical processes.

The marine biosphere acts as a reservoir for those elements that are selectively removed from sea water by organisms. In the case of sedentary populations this can provide local concentrations of elements. For example sessile invertebrates are a fixed reservoir for such elements as copper, zinc and cobalt which they concentrate by large factors. This will hold up the circulation of these elements, thus making their flushing rate from estuaries, for example, slower than that of the water.

Invertebrates growing on waste disposal containers, on the bottom, out of which radioactive elements are leaching may take up essentially all of such materials, the radioisotopes being effectively diluted only by the corresponding non-radioactive species in the water moving past the containers on the bottom. Such invertebrates can be eaten by commercial fishes and thus enter man's food. Calculations for at least some situations on the Pacific Coast indicate that the concentration of radioactive materials in foodfish under these circumstances can be much greater than if the material had been rapidly liberated into the overlying waters where quicker dilution by physical processes takes place.

As noted by KETCHUM[10], the combination of uptake by organisms in surface waters, and gravity effects on their excreta and dead bodies, will, in areas with positive estuarine type circulation, and regions of coastal upwelling, result in local concentrations of elements selectively taken up by such organisms. Even in the open sea, uptake by organisms may greatly impede the transfer to deep water of elements in particulate form which would be expected to settle out by sedimentation. For example HARLEY[11] found that cerium-144, which occurs in sea water in particulate form, and its daughter Pr^{144}, constituted from 80 to 90 per cent of fission product activity in plankton samples in the Western Pacific, after a considerable time following introduction into the sea.

Migrations of organisms can also have important effects in transporting elements from areas of high concentration to areas of low concentration. Vertical migrations of organisms, many of which readily cross the pycnocline, could readily transfer radioactive materials from the deep to surface waters. KETCHUM and BOWEN[7] have shown that this transport may in some circumstances be greater than that by physical means. Horizontal migrations can also transfer materials from the site of introduction to other areas. More important, however, is the fact that organisms used for food can be contaminated in one area and caught in distant uncontaminated areas. Some of the commercial fish species on our Pacific Coast, for example, make regular seasonal migrations to off-shore deeper waters and back to the shallower areas where they are caught. The sperm whale, which feeds on squids in the deep sea, but comes to the surface to breathe is an extreme example of a vertical migrant.

Present state of knowledge

Although our knowledge of most of these things is incomplete and fragmentary, we know

enough about the ecology of the sea to be able to make useful estimates of the quantities of radioactive materials that can be placed in various marine environments without hazard. This involves, however, making the most conservative (safest) assumptions wherever information is incomplete. The necessity of doing this means that waste disposal practices must be more restrictive than would be the case if we knew more. This, of course, entails economic costs which can in the future be very large, and which may be reduced by the results of further research. Research requirements have been outlined in several recent publications, including BROWN *et al.*[2], REVELLE and SCHAEFER[5] and SCHAEFER[6].

REFERENCES

1. R. REVELLE *et al*, *The Effects of Atomic Radiation on Oceanography and Fisheries*. Publication 551, National Academy of Sciences–National Research Council (1957).
2. H. BROWN *et al*, Oceanography 1960 to 1970. *Artificial Radioactivity in the Marine Environment* Chap. 5, pp. 1–31. National Academy of Sciences–National Research Council (1959).
3. D. F. CARRITT *et al*, *Radioactive Waste Disposal into Atlantic and Gulf Coastal Waters*. Publication 655, National Academy of Sciences–National Research Council (1959).
4. D. W. PRITCHARD *et al*, *Radioactive Waste Disposal from Nuclear Powered Ships*. Publication 658, National Academy of Sciences–National Research Council (1959).
5. R. REVELLE and M. B. SCHAEFER, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958 Vol. 18, pp. 364–370. United Nations, New York (1958).
6. M. B. SCHAEFER, New research required in support of radioactive waste disposal. *Proceedings of the International Conference on Radioactive Waste Disposal (Monaco), IAEA* (1960).
7. B. H. KETCHUM and V. T. BOWEN, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958 Vol. 18, pp. 429–433. United Nations, New York (1958).
8. R. REVELLE *et al*, *Report of the Committee on Oceanography and Fisheries*. The Biological Effects of Atomic Radiation, Summary Reports, pp. 69–90. National Academy of Sciences–National Research Council (1960).
9. *Report of the National Committee on Radiation Protection and Measurements, Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposures*, National Bureau of Standards, Handbook 69 (1959). Superintendent of Documents, Washington 25, D.C.
10. B. H. KETCHUM, *The Effects of the Ecological System on the Transport of Elements in the Sea*. Effects of Atomic Radiation on Oceanography and Fisheries, Publ. 551, pp. 52–57 (1957).
11. J. E. HARLEY (Editor), *Operation Troll*. U.S. Atomic Energy Commission, New York Operations Office, N.Y. (1956).

Health Physics Pergamon Press 1961. Vol. 6, pp. 103–109. Printed in Northern Ireland

DISPOSAL OF RADIOACTIVE WASTES IN THE OCEAN*†

DONALD W. PRITCHARD
Johns Hopkins University, Baltimore, Maryland

(*Received* 10 *October* 1960)

Abstract—An unavoidable consequence of the utilization of atomic energy for power production and other peaceful purposes is the introduction of some radioactive wastes into man's environment. The sea, as one segment of man's environment, will, both purposely and accidentally, receive some of these materials. It is the responsibility of all concerned to assure that such introduction will not endanger man or man's use of the resources of the sea. The purpose of this presentation is to outline the oceanographic problems associated with the peaceful uses of nuclear energy, to indicate what is now known concerning the various pertinent physical, chemical, biological and geological phenomena in the sea which determine the safe capacity of any segment of the sea to receive nuclear materials, and to present a preliminary evaluation of the capacity of the ocean to receive nuclear materials safely.

INTRODUCTION

In order to gain the benefits offered through the peaceful uses of nuclear energy and through the use of radioisotopes in medicine and in research, we must accept the fact that some artificially produced radionuclides will enter man's environment. The radiation dose which the human population will be subject to, over and above that imposed by the natural background, must be kept below levels which, on the basis of available evidence, will not impose undue risk to man. It is not only inconsistent but impossible to isolate one segment of man's environment and to prohibit all activity from entering that segment. Our basic responsibility is to utilize all segments of man's environment in a manner consistent with both the safety of the human population and the maximum beneficial use of nuclear energy. Inherent in the requirement of human safety is the requirement that man's use of products from any segment of his environment be not restricted in any way. The ocean, then, as one segment of man's environment, will be utilized as a receiver of nuclear materials. This symposium deals with the question of how such introduction can be made without undue risks to man or to his utilization of the marine environment.

MAJOR SOURCES OF WASTE MATERIALS ENTERING THE SEA

On the basis of available information from the various nations developing nuclear power and from public statements made at international meetings, it appears that radioactive materials now enter the sea in the form of packaged solids and liquids, as unconfined liquid effluent, and as unpackaged solid parts of contaminated equipment.

At the present time the major source of radioactive materials now being introduced into the sea, excluding fallout, is the low-level liquid effluent from spent fuel processing plants located on the sea coast or on rivers discharging to the sea. Packaged wastes contribute a very minor portion of the total activity entering the oceans. In the future an important source of radioactive materials entering the sea will be low-level liquid effluent and medium-level ion-exchange resins from nuclear-powered ships.

At present both fission products and materials having an induced activity enter the marine

* Presented at the Symposium on Waste Disposal, Fifth Annual Meeting of the Health Physics Society, Boston, Massachusetts, June 1960.

† Contribution No. 50 of the Chesapeake Bay Institute. This work was supported in part by the United States Atomic Energy Commission.

environment. It is to be expected that with improvements in reactor design and fuel processing procedures an even higher percentage of the fission products will be isolated from man's environment than is now the case, and materials having an induced activity will very likely become increasingly important as a component of the wastes which do enter the marine environment. An analysis of the waste disposal problems associated with nuclear-powered ships indicates that the corrosion products having an induced activity are by far the most significant component of the waste materials which might enter the sea from this source.

The fission product which has received the most attention from the health standpoint on land is Sr^{90}, which is much less a hazard in the sea because of the relatively large natural concentration of inactive strontium in marine waters. On the other hand, such radioactive materials as Co^{60}, Zn^{65}, Fe^{55} and Fe^{59}, Cr^{51}, and P^{32}, which occur as corrosion products in water-moderated reactors or in waste materials from research use of radioisotopes, may become the most significant components of waste materials entering the marine environment, since sea water has relatively small concentrations of the stable forms of these isotopes.

RELATIVE CONTAMINATION OF MARINE AND TERRESTRIAL ENVIRONMENTS

Excluding medical sources, the most likely route whereby radioactive materials originating with the peaceful use of nuclear energy will affect man is through the ingestion of food and water. This is certainly the source most important from the standpoint of presently occurring fallout products from weapons tests, and the situation should be similar for materials which enter man's environment through peaceful development of nuclear power.

At the present time food products from the sea have far lower radioactive content originating from fallout materials than do terrestrial food products. Considering the typical diet, marine food products contribute less than $\frac{1}{1000}$th of the total intake of radioisotopes by the average resident in the U.S.A., and about $\frac{1}{200}$th of the dietary intake of radioisotopes even for a seafood-eating nation such as Japan. This is in spite of the fact that general fallout has deposited about 2 Mc of Sr^{90} into the sea, and an additional 3 Mc of Sr^{90} have entered the Pacific Ocean as close-in fallout, as compared to about 1.8 Mc which have fallen on land. The two features of the marine environment which are of importance in this regard are the physical dilution of the isotope by vertical mixing and the isotopic dilution due to the presence in sea water of a relatively large concentration of inactive strontium.

EVALUATION OF THE SUITABILITY OF A MARINE LOCALE TO RECEIVE NUCLEAR WASTES

The fate of radioactive material introduced into the marine environment is dependent upon the following considerations:

(1) The physical and chemical form in which the material occurs at time of introduction, together with any relatively rapid changes in the physical and chemical character of the introduced material which occur when it is brought into contact with sea water. The subsequent dispersion by physical processes and reconcentration by the biota and bottom materials will be greatly affected by the physical and chemical form in which the wastes occur in sea water.

(2) Initial mechanical dilution of the wastes by the receiving waters, which will depend upon the manner of introduction. Thus a liquid waste will be subject to greater initial mechanical dilution if introduced as a strong jet into the body of the receiving waters than if introduced as a gently flowing stream on the surface. Large initial mechanical dilution is important in reducing the density difference between the initial contaminated volume and the surrounding receiving waters. This reduction in turn favors subsequent turbulent diffusion.

(3) Advection of the waste material away from the source region, and simultaneous turbulent diffusion, which lead to reduced concentrations of the radioactive components in the water.

(4) Uptake of the activity on to suspended silt and bottom sediments, which removes some

of the radioactive materials from the water, and restricts further dispersion. In deep water such removal would be favorable since material incorporated with the bottom sediments would be unlikely to return to man's environment. In shallow coastal areas containing bottom-living shellfish and bottom-feeding commercial fin fish, concentration of radioactivity on the bottom may be unfavorable since these detritus and filter feeders may further concentrate the activity from the bottom material.

(5) Concentration of activity by various parts of the biota, including shellfish and fin fish important to man as a source of food. Some important fission products and corrosion products are concentrated by marine organisms by factors of from 100 to 10,000 over the concentrations of these isotopes in the water.

The evaluation of the suitability of any particular marine locality as a receiver of nuclear wastes ideally involves the precise, step-by-step consideration of all factors affecting the possible return of radioactive material to man. The general procedure is the same whether the evaluation concerns: (a) the selection of suitable disposal areas for packaged wastes; (b) the selection of the position of an outfall discharging low-level liquid effluent from a chemical processing plant; (c) the consideration of the suitability of a given harbor or harbor approach to receive low-level liquid wastes from nuclear powered ships; or (d) the determination of the suitability of the mixed layer of the open ocean to receive wastes from the ion exchange resins on nuclear powered ships.

Fig. 1 presents in schematic form such a step-by-step procedure. The solid arrows between blocks on the diagram indicate the route taken by the radioactive material in returning to man, while the dashed arrows indicate the reverse course taken in the evaluation. The starting point in the evaluation is *man*.

To evaluate the problem, a maximum permissible rate of exposure must be adopted. The usual basis of such a maximum exposure rate is that recommended for the population-at-large by the national and international committees on internal dose.[1,2]

Considerable care should be taken that the potential additive effect of exposure to radiation from various sources is given due consideration. The second step in the evaluation is then a consideration of all sources of radiation exposure to the particular segment of the population which would also be potentially affected by the waste disposal operations. On the basis of such consideration a portion of the maximum permissible dose is assigned to sea disposal.

The next step is to consider the various routes by which radioactivity can reach man from the marine environment.

The possible danger of direct radiation, the harvest of seafood, possible contamination of fishing gear and of beach sand are some of the routes which should be considered at this stage of the study.

The determination of maximum permissible concentrations of various isotopes in those parts of the marine environment (seafood, bottom sediments, and shore material) which constitute routes by which radioactivity may return to man from the sea then follows as the next step in the process. From consideration of known factors by which the biota and the sediment concentrate various radioactive isotopes from sea water, it is then possible to arrive at the maximum permissible concentration of the various isotopes in the sea water.

The final steps involve evaluating the changes in concentration and distribution of radioactivity which may be brought about by:

(1) Exchange between bottom sediment and suspended or dissolved material in sea water.

(2) Advection and turbulent diffusion, both within a given marine environment and between adjacent environments.

(3) Initial mechanical dilution, influenced by manner of discharge.

(4) Physical and chemical form of the wastes released.

The end result is an estimate of the maximum rate of release which will not exceed the maximum permissible concentration in sea water.

The marine biologist and the oceanographer are essentially concerned with those steps dealing with (a) concentration factors from sea water to the various parts of the marine environment which constitute the return routes to man (i.e. the food chain, beach sand, etc.); (b) rate of transfer of radioactive materials to

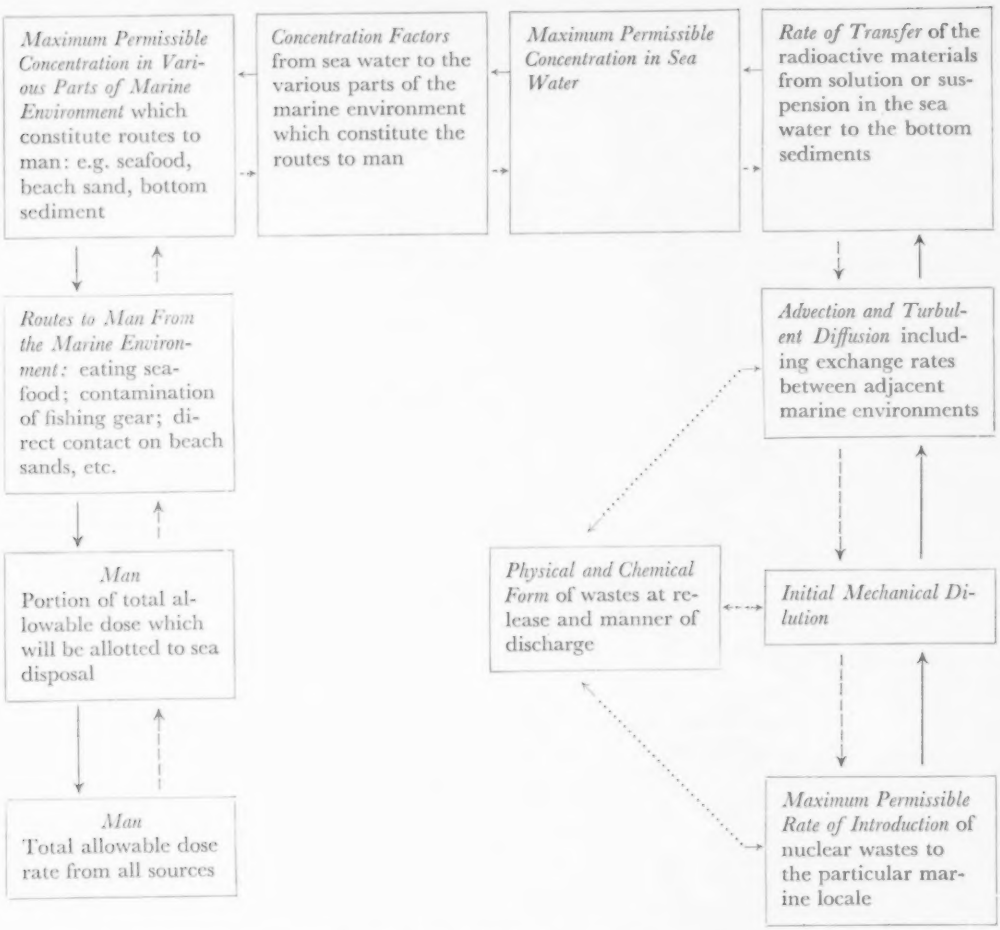

Maximum Permissible Concentration in Various Parts of Marine Environment which constitute routes to man: e.g. seafood, beach sand, bottom sediment
Concentration Factors from sea water to the various parts of the marine environment which constitute the routes to man
Maximum Permissible Concentration in Sea Water
Rate of Transfer of the radioactive materials from solution or suspension in the sea water to the bottom sediments
Routes to Man From the Marine Environment: eating seafood; contamination of fishing gear; direct contact on beach sands, etc.
Advection and Turbulent Diffusion including exchange rates between adjacent marine environments
Man
Portion of total allowable dose which will be allotted to sea disposal
Physical and Chemical Form of wastes at release and manner of discharge
Initial Mechanical Dilution
Man
Total allowable dose rate from all sources
Maximum Permissible Rate of Introduction of nuclear wastes to the particular marine locale

FIG. 1. Schematic presentation of the step by step considerations which should be made in evaluating the suitability of any marine locale as a receiver of nuclear wastes.

bottom sediments from solution or suspension in the sea water; (c) advection and turbulent diffusion of the introduced contaminant; and (d) the effects of the physical and chemical form of the wastes, and the manner of discharge.

Understanding of oceanographic aspects of the problem is far from complete, and considerable research is indicated if reasonably precise estimates are to be made of the capacity of the marine environment to receive nuclear waste material. However, oceanographic knowledge is presently sufficient to permit order of magnitude estimates of the rate at which nuclear materials may be introduced to the sea without undue risk to man. Present knowledge is sufficient to assure that such estimates will be conservative—that is, on the safe side.

MAXIMUM PERMISSIBLE CONCENTRATIONS OF RADIOACTIVE ISOTOPES IN SEA WATER

In the present state of knowledge, the permissible concentrations in sea water can be regarded only as interim values; moreover,

they should be thought of as average values for the relatively large volumes of water from which edible marine animals and plants extract their body materials.

In estimating permissible concentrations in sea water, account must be taken of the fact that sea water is a solution of almost all the elements; that the concentrations of many elements are known and are constant within more or less well-defined limits; and that marine plants and animals concentrate, often by very large factors, both radioactive and non-radioactive isotopes.

Hazards to human beings from a radioactive nuclide in seafood will result from radiation in the gastrointestinal tract and from radiation in other body organs which accumulate the radionuclide. As shown in National Bureau of Standards Handbook 69[2] the maximum permissible concentrations in drinking water for many isotopes, including Cr^{51}, Fe^{59}, Co^{60}, Ru^{106}, Ce^{144} and Ta^{182}, are limited by the radiation exposure of the gastrointestinal tract; for others, including P^{32}, S^{35}, Ca^{45}, Fe^{55}, Zn^{65}, Sr^{90}, I^{131} and Cs^{137}, the accumulated burden in body organs is limiting.

For the first group of isotopes, the concentration per unit volume of marine food must be held below a certain value; for the second group, the specific activity (that is, the ratio of the radioactive to the non-radioactive species in the seafood) must be controlled.

Organisms do not, in general, distinguish between the radioactive and the non-radioactive isotopes of a particular element. Hence, if the total uptake of any element by a human body organ comes from eating seafood, the specific activity in the body cannot, in general, exceed the specific activity in the ocean. (Exceptions may possibly occur if the radioactive and the non-radioactive species are in different chemical states in sea water.) Indeed, the specific activity of the radioactive isotope accumulating in the body will be much less than in the ocean if the isotope has a short radioactive half-life, because the concentration in marine food organisms and in body organs will be reduced by radioactive decay. Consequently, whenever the gastrointestinal tract is not the critical body organ, the permissible specific activity of a radioisotope in sea water will be greater than the permissible specific activity in the body, often by a large factor for isotopes with a short radioactive half-life. (Obviously, this will also be true when the concentration in sea water of the stable species of the element is very small. In these cases, the ratio of the radionuclide to a non-isotopic carrier used by the body must be considered.)

Table 1. *Comparison of permissible sea water concentrations (tentative) with one-tenth of the MPC values for drinking water from Handbook* 69[2,3]

Isotope	One-tenth MPC for drinking water (μc/ml)	Tentative permissible sea water concentrations (μc/ml)
P^{32}	2×10^{-5}	4×10^{-9}
S^{35}	6×10^{-5}	1×10^{-3}
Ca^{45}	9×10^{-6}	1×10^{-4}
Cr^{51}	2×10^{-3}	2×10^{-5}
Fe^{55}	8×10^{-4}	1×10^{-6}
Fe^{59}	6×10^{-5}	6×10^{-9}
Co^{60}	5×10^{-5}	5×10^{-8}
Zn^{65}	1×10^{-4}	7×10^{-9}
Sr^{90}	1×10^{-7}	3×10^{-6}
Nb^{95}	1×10^{-4}	5×10^{-6}
Ru^{106}	1×10^{-5}	1×10^{-7}
I^{131}	2×10^{-6}	2×10^{-6}
Cs^{137}	2×10^{-5}	1×10^{-7}
Ce^{144}	1×10^{-5}	1×10^{-8}

The National Academy of Sciences[3] Committee on the Effects of Atomic Radiation on Oceanography and Fisheries has published a table of tentative permissible sea water concentrations of a number of radionuclides. The material contained in Table 1 is drawn from that source.

It will be noted that the permissible sea water concentrations for S^{35}, Ca^{45}, and Sr^{90} are larger by factors of 15 to 30 than the maximum permissible concentrations for drinking water. The stable isotopes of these elements are present in relatively large amounts in the oceans, and they are not greatly concentrated by marine organisms. On the other hand, such substances as P^{32}, Fe^{59}, Co^{60}, Zn^{65}, and Ce^{144} have permissible sea water concentrations from $\frac{1}{1000}$th to

$\frac{1}{10,000}$th of the maximum permissible concentrations for drinking water. Here, the quantities of the stable isotopes in sea water are quite small, and these elements are heavily concentrated by marine organisms.

ALLOWABLE DISPOSAL RATES INTO THE SEA

Using estimates of the permissible sea water concentrations, and taking conservative estimates regarding the availability of the introduced material to the marine biota, the rate of diffusion of the material from the source, and such other steps involved in the return of the activity to man, it is possible to compute the rate at which a particular marine locale can receive nuclear wastes without introducing an undue risk to man.

For use as a receiver of packaged wastes, consider for example a disposal area off the coast of the United States, having an area of 10 by 10 miles, and located in depths greater than the lower limit of the layer of primary production (about 50 m). Let us make the conservative assumptions that: (1) no radioactive decay takes place between the time of disposal and the time of return to man in seafood; (2) the radioactive material is immediately released from the container and taken up by sessile organisms growing on the container; (3) edible fish which consume these sessile organisms live all their lives in the disposal area; (4) only the lower 2 m of the water column are available for dilution; (5) the flow of water through the disposal site is at the relatively slow rate of 1 km/day; (6) man receives all his protein requirements from such fish. We arrive at the relationship: allowable disposal rate for a single isotope, in curies per year, is equal to 1.2×10^{10} times the permissible concentration of the isotope in sea water expressed in microcuries per milliliter.

If the disposal site is in water having depths greater than the deepest depth at which food fish for man live, then, under the most restrictive assumptions regarding diffusion, the above factor of 1.2×10^{10} would be multiplied by a factor equal to the depth interval, in meters, between the greatest depth of food fish production and the bottom depth. Our best information is that food fish do not occur below 2000 m. If the disposal site were 4000 m deep the relationship would be: allowable disposal rate for a single isotope, in curies per year, is equal to 2.4×10^{13} times the permissible concentration of the isotope in sea water, expressed in microcuries per milliliter.

For a mixture of isotopes, the allowable rate for each would be reduced to take into account the additive effects.

Table 2 lists the allowable disposal rates, in curies per year, computed for these two examples. Though these rates are based on tentative permissible sea water concentrations, the multiplicity of conservative assumptions assure that these results are also certainly conservative.

Table 2. Example of computed allowable rates of disposal of specific isotopes, in curies per year, for a sea disposal site of 10 miles *by* 10 miles *dimension, and which is: (A) in water depths less than the limit to which food fish inhabit; and (B) in water depths* 2000 m *deeper than food fish inhabit*

Isotope	Disposal rate case *A*	Disposal rate case *B*
P^{32}	48	96×10^3
S^{35}	12×10^6	24×10^9
Cr^{51}	24×10^4	48×10^7
Fe^{55}	12×10^3	24×10^6
Fe^{59}	72	14×10^4
Co^{60}	600	12×10^5
Zn^{65}	84	16×10^4
Sr^{90}	36×10^3	72×10^6
I^{131}	24×10^3	48×10^6
Cs^{137}	12×10^2	24×10^5

These disposal rates are particularly conservative for isotopes such as P^{32} and I^{131} which have relatively short radioactive half-lives and relatively long biological half-lives.

NATIONAL AND INTERNATIONAL ACTIVITIES

About 5 years ago the National Academy of Sciences undertook a study of the biological effects of atomic radiation. One of the six committees under this study is the Committee on the Effects of Atomic Radiation on Oceanography and Fisheries, known in governmentese as CEAROF. Working panels of this committee

have prepared reports on packaged waste disposal on the Atlantic and Gulf coasts of the United States, and also on waste disposal from nuclear-powered ships. A third working panel is now completing a report on packaged waste disposal on the Pacific Coast.

In the fall of 1958 the International Atomic Energy Agency in Vienna appointed a panel on the problems of radioactive waste disposal into the sea. The final report of this panel has just been issued by the IAEA and contains recommendations regarding sea disposal and the international monitoring of sea disposal operations. To date all serious and reasonably complete studies of the problem of radioactive waste disposal to the sea have concluded that the oceans can be utilized to receive at least limited amounts of radioactive materials.

REFERENCES

1. *Recommendations of the International Commission on Radiological Protection, Report of Committee II on Permissible Dose for Internal Radiation,* Pergamon Press, London (1959); *Health Phys.* **3** (1960).
2. *Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and in Water for Occupational Exposure,* National Bureau of Standards Handbook 69. Superintendent of Documents, Washington, D.C. (1959).
3. *Report of the Committee on the Effects of Atomic Radiation on Oceanography and Fisheries.* In *The Biological Effects of Atomic Radiation,* Summary Reports p. 83. National Academy of Sciences–National Research Council (1960).

Health Physics Pergamon Press 1961. Vol. 6, pp. 110–113. Printed in Northern Ireland

SOME LEGAL CONSIDERATIONS IN THE OCEAN DISPOSAL OF RADIOACTIVE WASTES*

REMARKS

ROBERT LOWENSTEIN
Counsel, Division of Licensing and Regulation, Office of the General Counsel,
U.S. Atomic Energy Commission

(*Received* 10 *October* 1960)

I AM very happy to be invited once again to participate at the annual meeting of the Health Physics Society. At the time of my first appearance before you, at your meeting in Columbus, Ohio, in 1955, I was asked to discuss the regulatory provisions of the newly enacted Atomic Energy Act of 1954, and the program which the Commission was developing to implement the Act.

In the 5 years since then, legal problems which we then envisaged as being far in the future—and also some we did not anticipate—have actually come to confront us. There has been litigation as to the proper criteria for licensing power reactors and the ocean disposal of radioactive wastes; there have been actions for personal injuries resulting from radiation exposures; new insurance pools with a total domestic capacity of $60 million have been organized to insure against liability for damages from nuclear incidents; indemnity legislation has been enacted to protect against the remote possibility of catastrophic nuclear incidents; legislation has been enacted to provide a greater role for the States in controlling atomic energy hazards; many administrative proceedings have been conducted before the Atomic Energy Commission's hearing examiner in connection with the issuance, suspension, revocation or modification of licenses. Work is under way to improve workmen's compensation laws and to develop uniform rules of tort liability. Similar problems have arisen in foreign countries, and at the international level, and are being resolved along lines which, in most cases, are not fundamentally different from the approaches which have been adopted in the United States.

During these same years, the Commission's regulatory program has expanded. Since 1 January, 1955, the number of reactor construction permits and operating licenses has grown from 0 to over 75; byproduct material licensees from about 2750 to almost 5500; special nuclear material licenses from three to 294; source material licenses from three to almost 300. Many of the more fundamental standards and practices which you have developed, in your day-to-day activities, have in one form or another been adopted as legal requirements by incorporation in the regulations of State and local radiation control agencies as well as the regulations of the Atomic Energy Commission.

In carrying out its regulatory program under the Administrative Procedure Act and the Atomic Energy Act, the Atomic Energy Commission performs two fundamental kinds of functions. In performing quasi-legislative functions, under authority delegated by the Congress,

* Presented at the Symposium on Waste Disposal, Fifth Annual Meeting of the Health Physics Society, Boston, Massachusetts, June, 1960.

the Commission establishes the basic rules for protection of health and safety from uses of AEC-licensed materials. When published as regulations, these rules have essentially the same legal effect as statutes enacted by the Congress. In addition to rule-making, the Commission performs quasi-judicial or "adjudicatory" functions to determine whether or not in the specific case the "rules" are being observed. Proceedings for the issuance, modification or revocation of a license are typical of the Commission's adjudicatory functions. Of course, in practice, the functions of rule-making and adjudication frequently tend to merge, particularly in situations where, as with regard to the ocean disposal of radioactive material, the published regulations establish broad criteria rather than specific requirements.

There are presently about nine licenses in effect authorizing the ocean disposal of radioactive wastes. Adjudicatory hearings have been held in three cases (Matter of Walker Trucking Company, Docket no. 27–5; Matter of Coastwise Marine Disposal Company, Docket no. 27–13; and Matter of Industrial Waste Disposal Corporation, Docket no. 27–9). Since the recent Gulf of Mexico case (Matter of Industrial Waste Disposal Corporation) presents issues typical of these proceedings, and was decided by the Commission only last week, it seems most apt to discuss the principal legal problems of ocean disposal by reference to that case.

The application in Industrial Waste Disposal Corporation was filed in 1958. Because of the considerable public interest, the Commission set the application down for hearing in Houston, Texas, in January, 1959. At the hearing, the City of Corpus Christi, the Nueces County Navigation District, the County of Nueces, the Sportmen's Clubs of Texas, Inc., and Harris County, Texas, were granted leave to intervene. A representative of the Government of Mexico attended the hearing but did not participate other than to inquire of one witness as to the extent of oceanographic research at the proposed disposal site. The Presiding Officier rendered a decision on 25 May, 1959, in which he directed that the license be issued. The intervenors filed exceptions to his decision, requesting review of the decision by the full Commission. The State of Texas then also requested and was granted permission to intervene and file exceptions.

During the summer of 1959, the Government of Mexico raised questions concerning the proposed license to dispose of waste in the Gulf of Mexico. In a note dated 15 June, 1959, to the U.S. State Department, the Mexican Government expressed its

> . . . regrets that the Atomic Energy Commission of the United States continues to insist on its original project which, it appears, will result in permitting the firm, Industrial Waste Disposal Corporation of Houston, Texas, being authorized from June 20, 1959, to dispose of radioactive wastes in the Gulf of Mexico at equal distances from the coast of Mexico and the coast of Texas.
>
> The Government of Mexico, for reasons of a technical and scientific nature expressed previously, as well as because of the unfavorable impression that the project can have on Mexican public opinion, is obliged to deposit its expression of disagreement in this matter.

The Department of State forwarded the Mexican note to the Atomic Energy Commission under a memorandum which advised the Commission, among other things, that:

> From the note of the Mexican Embassy it may be inferred that the Mexican Government believes disposal of radioactive wastes so close to its shores would be interpreted in Mexico as a unilateral and arbitrary act on the part of the United States, any adverse results from which would present virtually identical hazards to the residents of the two countries.
>
> Consequently, the Department believes that the granting of the license application would have seriously harmful effects on our friendly relations with Mexico and with the other countries of the hemisphere, since most of them would surely agree with Mexico that such an act would be arbitrary.

To permit resolution through the State Department of the objections of the Government of Mexico, argument on the exceptions was postponed first to December 1959 and then to January 1960.

In its decision, the Commission directed that the license be issued to the extent of authorizing the applicant to receive radioactive waste materials; to store them at the applicant's storage site in Houston; and to transport them

to Commission-operated disposal sites within the United States. The Commission remanded the case to the Hearing Examiner for further proceedings to take further evidence concerning the integrity of the types of containers described in the application after disposal in the Gulf of Mexico, and such other evidence as the Hearing Examiner may allow.

As the case was briefed and argued before the Commission, there were four principal issues:

1. *What weight should be given to the Department of State communication concerning the objections of the Government of Mexico?*

The AEC staff recommended that the Commission deny the license application, without regard to the technical merits of the application and without prejudice to renewal of the application upon withdrawal of the Mexican objections.

In support of this position, the staff argued that the Commission is obliged to consider foreign affairs questions when properly raised in licensing proceedings before the Commission. This argument was made on the basis that the Atomic Energy Act of 1954 requires the Commission to consider pertinent questions of the "common defense and security", as well as public health and safety; and that the Atomic Energy Commission as an agency in the executive branch of the Government is obliged to act in a manner consistent with the inherent authority of the President under the Constitution to conduct the foreign affairs of the United States. The staff urged also that, under the circumstances of this case, the Commission was obliged to accept the views of the Secretary of State, as the President's spokesman, with respect to the impact on our foreign affairs of issuance of the proposed license.

The Commission's decision did not consider the foreign affairs question because the case was remanded to the hearing examiner for other reasons.

2. *The applicability of a City of Houston ordinance prohibiting the operation of any radioactive waste disposal plant within the city*

Intervenors urged that the Commission should not issue the license unless there was compliance with applicable ordinances of the City of Houston. The AEC staff urged, and the Commission decided, that the applicability and validity of the ordinance are not proper questions for consideration by the Commission in a licensing proceeding. Such laws frequently raise legal questions (such as questions as to the proper interpretation of the laws, their validity under the State constitution, etc.) which should be decided by the regular courts and are not within the competence or jurisdiction of the Commission.

3. *Intervenors raised issues as to the applicant's qualifications to engage in the proposed activities and the suitability of his proposed waste, storage and handling site*

The Commission found that these, as well as the applicant's proposed equipment and procedures for handling the waste, would be adequate.

4. *The issue on which the Commission was not satisfied concerned proof as to the integrity of the containers*

The applicant proposed over the 2-year period for which he requested a license, to dispose of a total of not more than 240 c of activity in barrels meeting ICC specifications and containing not more than 2 c in each barrel. The containers would be packaged in either of two ways:

(a) Where the container of radioactive waste received by the applicant from his customers is smaller than a 55-gal drum, it would be centered in the drum, surrounded by concrete reinforced with wire mesh, and the drum would be closed by means of a bolted, steel, closing ring;

(b) Where the container itself is a 55-gal drum, it would be completely encased in a sheet of steel-reinforced concrete at least 5 in. thick.

No containers would be shipped to the applicant except 55-gal drums or containers small enough to fit in a 55-gal drum with a 5-in. coat of concrete. Pressure relief valves would not be required.

The case was decided by three Commissioners. The majority, consisting of Chairman JOHN A. MCCONE and Commissioner JOHN S. GRAHAM,

stated with respect to the container question:

> Our examination of the record does not disclose any comprehensive testimony concerning the effect of sea pressure, at various depths, upon the two types of containers proposed to be used by the Applicant. Although we recognize that the testimony of record may support a conclusion that, even if such a container ruptured, no unreasonable hazard to human and animal life would result, we feel constrained to take a conservative approach on the matter of container integrity and we believe it appropriate to require this Applicant, if he desires authorization to dispose at sea, to present further testimony with respect thereto on our remand of this proceeding.
>
> We believe that consideration should be given on the remand of this case to the admixture of waste with concrete, in a 55-gal drum so as to constitute a virtually solid block and thus to avoid any chance of implosion of the container at the authorized disposal depth. Such a procedure would appear to avoid the alleged problem of preventing significant voids when waste is packed either in 55-gal drums or in smaller containers which in turn are placed in concrete within such drums. Additionally, the Applicant may also wish to explore the use of a valve which would permit equalization of pressure in any voids as the barrel descends.

In a separate opinion, in which he concurred in the decision to remand, Commissioner JOHN F. FLOBERG stated:

> I consider this decision to be principally based on grounds other than supportable scientific or technical conclusions that either the integrity of the containers or a change of site is essential or, indeed, even relevant to the health and safety of the residents of the land areas surrounding the Gulf of Mexico. Rather I believe it to be based on the eagerness of the Commission to allay all anxiety, however unreasonable, unfounded, and scientifically unsound, of the residents of those areas with regard to safety.
>
> That rupture of the disposal packages at any reasonable depth would pose no threat to public health and safety of the residents of either the United States or Mexico, I am completely satisfied.*

I would suppose that the overwhelming majority of you, as health physicists, have no doubt that the ocean disposal proposed by the applicant would be without hazard to health and safety. In atomic energy, however, as in all other activities in a democratic society, it is not enough merely to be confident we are right. No activity in a democratic country can be carried on for long without public approval. This need for public approval imposes upon those with specialized knowledge in areas of public concern, the responsibility to communicate that knowledge in understandable terms in order to create an informed public opinion.

Having worked quite closely with many of you for almost 10 years, I recognize, and I believe I understand, the impatience which many of you feel at becoming involved in legal proceedings, and your impatience with legal requirements of applications and licenses, with radiation regulations that establish exposure limits rather than guides.

Had uses of atomic energy remained in the laboratory where they were initially developed not very many years ago, you would not be so involved with things legal today. That you are so involved, demonstrates the success of efforts to develop uses of the atom. Having emerged from the laboratory, the atom, like other significant technologies, has become involved in social, economic and other controversy. The law and legal proceedings provide the standards and the means by which such controversy is resolved—they do not create the controversy. The law (substitute "regulations", if you wish) provides also the minimum requirements of safe practices from which users may not depart even when costs are mounting and economic competition is severe. Thus, these requirements protect you, whose profession is safety, and your organizations from the undermining effect of competition from organizations to whom safety might be of secondary importance.

* After the delivery of this talk, one of the intervenors in the Industrial Waste Disposal Corporation Case, Harris County, Texas, appealed the Commission's grant of the license to the federal courts. On June 30, 1961, the United States Court of Appeals for the Fifth Circuit upheld the Commission's authority over radioactive waste disposal and its action in this case.

Health Physics Pergamon Press 1961. Vol. 6, pp. 114–125. Printed in Northern Ireland

PERMISSIBLE CONCENTRATIONS OF RADIONUCLIDES IN SEA WATER

A. H. W. ATEN, JR.
Institute for Nuclear Physics Research, Amsterdam, Netherlands*

(*Received* 10 *October* 1960)

Abstract—Methods are discussed for the calculation of the maximum permissible concentration of various radionuclides in sea water. The values are based on the maximum permissible concentrations in drinking water as fixed by the I.C.R.P. Concentration factors of radionuclides in fish are discussed and the protection of fish against the uptake of radioactive nuclides by inactive isotopes and by elements in the sea water which are chemically related to the radioisotopes is taken into account.

In the Appendix the application of the maximum permissible concentrations in sea water is discussed for simplified case of waste disposal into the sea.

INTRODUCTION

For discussions concerning the amount of radioactivity that can be dumped into various seas or oceans it is convenient to introduce the notion of a "maximum permissible concentration in sea water" $(MPC)_s$.† This quantity indicates the concentration of a certain radioactive nuclide which, if uniformly present in a certain sea, would result in a concentration of this radioactive nuclide in the fish which would cause anyone who consumed this fish (in an amount equal to the maximum daily consumption) to have a daily uptake of the radioactive nuclide equal to the maximum permissible uptake. Of course, if more than one radioactive nuclide were present the permissible concentrations would be lower, just as is the case in drinking water. Also it is possible that not all the permissible radioactivity in the human body may be supplied by eating contaminated fish which would mean that the permissible concentration in sea water would be lowered by an extra factor. Evidently, the assumption that the radioactivity is distributed homogeneously throughout the sea is not a realistic one, but even so the notion of $(MPC)_s$ will be found useful in discussing the quantity of radioactivity which can be tolerated in a certain sea as we shall demonstrate in the Appendix to this report.

The recent publication of new recommendations by the International Commission on Radiological Protection[3,4] makes it possible to estimate the permissible concentrations in sea water of those radionuclides which play an important role in the technology of radioactivity. As such we shall consider the nuclides listed in Table 1. (This table also indicates the maximum permissible concentrations for the population at large based on the gastrointestinal tract and an alternative most sensitive organ.) The list is, of course, fairly arbitrary at this stage.

BASIC DATA

The maximum permissible concentrations in sea water in this paper will be based on the amount of radioactivity which may be taken up from fish. (If the consumption of other marine animals does not exceed 25 g/person as an average over periods of months, the limitation imposed by their consumption does not for most nuclides impose lower tolerances

* This investigation was performed as part of the programme of the Foundation for Fundamental Research of Matter (F.O.M.) with financial support of the Organization for Pure Research (Z.W.O.).

† Several of the basic notions used in this paper, can also be found in two recent reports of the National Research Council on the disposal of radioactive waste in sea water.[1,2] The quantity $(MPC)_s$ corresponds roughly to PSC in Ref. (1), although the two differ by small numerical factors.

Table 1. *Radionuclides which are important in atomic energy*

Nuclide	Half life	First limiting organ*	$(MPC)_w$ (μc/ml)	Second limiting organ*	$(MPC)_w$ (μc/ml)
Fe^{59}	45 days	G.I.	2×10^{-5}	total body	2×10^{-5}
Co^{58}	71 days	G.I.	3×10^{-5}	total body	4×10^{-5}
Co^{60}	5.3 years	total body	10^{-5}	G.I.	1.7×10^{-5}
Zn^{65}	245 days	total body	10^{-5}	G.I.	6.7×10^{-5}
Sr^{89}	51 days	bone	3×10^{-6}	G.I.	1.3×10^{-5}
Sr^{90}	28 years	bone	3×10^{-8}	G.I.	1.7×10^{-5}
Y^{91}	58 days	G.I.	10^{-5}	bone	10^{-2}
Zr^{95}	65 days	G.I.	2×10^{-5}	total body	0.01
Nb^{95}	36 days	G.I.	3×10^{-5}	total body	0.04
Ru^{103}	40 days	G.I.	2.7×10^{-5}	total body	8×10^{-4}
Ru^{106}	1.0 year	G.I.	3×10^{-6}	kidney	1.3×10^{-4}
I^{131}	8.1 days	thyroid	6.7×10^{-7}	G.I.	3×10^{-4}
Cs^{137}	30 years	total body	2×10^{-6}	G.I.	2.7×10^{-4}
Ce^{141}	33 days	G.I.	3×10^{-5}	liver	0.1
Ce^{144}	285 days	G.I.	3×10^{-6}	bone	2.7×10^{-3}
Pr^{143}	14 days	G.I.	1.7×10^{-5}	total body	0.1
Pm^{147}	2.6 years	G.I.	6.7×10^{-5}	bone	1.7×10^{-2}
Sm^{151}	93 years	G.I.	1.3×10^{-4}	bone	1.7×10^{-2}
Ra^{226}	1625 years	total body	2×10^{-9}	G.I.	1.7×10^{-5}
Th^{230}(Io)	8×10^4 years	bone	6.7×10^{-7}	G.I.	10^{-5}
U nat	very long	G.I.	6.7×10^{-6}	kidney	2×10^{-5}
U^{235}	7×10^8 years	G.I.	10^{-5}	total body	10^{-4}
Pu^{239}	2.4×10^4 years	bone	1.7×10^{-6}	G.I.	10^{-5}

* One of the two limiting organs for every nuclide has been taken to be the G.I. This does *not* mean that there may not be more than one other organ more sensitive to the radiation of the nuclide than the G.I.

than the consumption of fish.) We shall assume that the maximum amount of fish consumed per day amounts to 250 g.[5] In our calculation of $(MPC)_s$ we do not at this stage introduce a factor to take into account the dilution of contaminated fish by fish from other regions and we shall, at least to begin with, aim at a maximum uptake of radioactivity by any person not exceeding the daily dose permitted by the I.C.R.P. *for the population at large*. This means that we shall base our estimates on the maximum permissible concentrations for drinking water† suggested by the I.C.R.P. for occupational exposure multiplied by the safety factors indicated in Table 2. Furthermore we shall assume—as is done in the I.C.R.P. report—that only one single radioactive species is consumed. In case more than one type of radioactive element is consumed the sum of the fractions of the maximum permissible concentrations for the different nuclides should not exceed unity.

One of the most important data needed to estimate MPC-values for sea water is the concentration factor (F_c) which indicates the ratio of the concentrations of the radionuclide in fish and in sea water.‡ In case the concentration factor is known with sufficient accuracy we can calculate the maximum permissible concentration in sea water $(MPC)_s$ from the maximum

Table 2. *Safety factors used to convert occupational* $(MPC)_w$ *values to corresponding figures for the population at large*

Safety factor	$\frac{1}{30}$	$\frac{1}{100}$
Critical organs	gastrointestinal tract bone thyroid kidney liver	total body

† All $(MPC)_w$ values used in this report refer to a 168-hr week. They differ from the figures listed in Ref. (4), because they have been obtained by multiplying the latter by the safety factors in Table 2.

‡ The symbol F_c corresponds to the factor f in Refs. (1) and (2).

Table 3. Concentration factors in fish

Element	F_c observed	Method
Potassium	5	Chemical analysis[6]
	10	Chemical analysis[7]
	best value 8	
Calcium	1	Chemical analysis[6]
	22	From concentration in sea water[8]
	best value 2	
Titanium	40	Chemical analysis (very inaccurate)[6]
Iron	1000	Chemical analysis[6]
Cobalt	160	Chemical analysis of fish and sea water[6,9]
Zinc	1000	Chemical analysis[6]
	5000	Chemical analysis[7]
	best value 3000	
Strontium	1	Chemical analysis[6]
	1	Chemical analysis (and uptake?)[10]
	0.3	Uptake of Sr^{89} in *Tilapia*[11]
	>5	Uptake of Sr^{85} in mullets (whole fish)[12]
	0.25	Uptake of Sr^{85} in killifish (muscle)[12]
	1.0	Uptake of Sr^{85} in young eel (equilibrium doubtful)*
	4	Chemical analysis fishflesh and uptake of Sr^{90} [13]
	best value 2.0	
Yttrium	5	Chemical analysis (and uptake?)[10]
Ruthenium	3.2	Calculated from comparison of uptake of Ru^{106} and Sr^{90} with $F_c = 2.0$ for strontium[5]
Iodine	10	Chemical analysis[6]
	3	Chemical analysis[7]
	~7	Small eel (uptake of I^{131})*
	best value 7	
Caesium†	10	Chemical analysis[6]
	7	Chemical analysis (and uptake?)[10]
	>5	Uptake of Cs^{137} in croakers (muscle) (extrapolation)[11]
	>35	Uptake of Cs^{137} in killifish (muscle)[12]
	~25	Extrapolated uptake of Cs^{137} in young eel*
	best value 20	
Cerium	12	Chemical analysis (and uptake?)[10]
Radium	15	Calculated from radium content of fish[7] (this is supported by determinations by MAYNEORD *et al.*[14] and of sea water[8,15]
Uranium	<20	Chemical analysis (meat of different of fish)‡
Plutonium	13	Calculated from comparison of uptake of Pu^{239} and Sr^{90} with $F_c = 2.0$ for strontium[5]

* Measurements by Miss G. WOLZAK and Miss J. C. KAPTEIJN obtained in this laboratory.

† There is also an observation[13] to indicate that F_c for caesium is four times the value for potassium, which is in good agreement with our choice of best values.

‡ Measurements performed by Dr. KOOI and Dr. U. HOLLSTEIN of Reactor Centrum Nederland and J. W. DALENBERG of this Institute and the author.

permissible concentration in drinking water $(MPC)_w$ by means of the following formula:

$$(MPC)_s = \frac{8.8}{F_c} \times (MPC)_w \qquad (1)$$

It is evident that it will be of the greatest importance to us to use the best values at our diposal for the concentration factors. The available information is discussed in Table 3. It is seen that the inaccuracy of even the (averaged) best values of the concentration factors is appreciable and in general not less than a factor amounting to several times unity. This is specially true for the concentration factors of ruthenium and plutonium calculated from DUNSTER's average (see Table 9 in Ref. 5) in which the figure for Sr^{90} is somewhat uncertain, due to the presence of Sr^{89}. This tends to make the F_c-values for Ru and Pu too low). Probably, concentration factors to a certain extent will depend on the kind and size of the fish, and possibly even on the other conditions in the sea, but even so we may try to use the notion of a constant concentration factor of an element in fish as a useful approximation.

Just as in the case of the drinking water tolerances the radionuclides in sea water can be arranged in two groups, one group which is limited by the danger of over irradiating the gastrointestinal tract (G.I.) and another group for which the limiting factor is the radiation dose administered to some other critical organ. It is easily seen that the division is the same in the case of maximum permissible concentrations in sea water as in drinking water.

HYDROLYSABLE ELEMENTS

It is of interest to point out the chemical meaning of these two groups. A number of elements hydrolyse quite easily, and these will in general be unable to leave the gastrointestinal tract, the latter for this reason being the limiting organ for radioisotopes of these elements. It is to be expected that the same elements will also tend to hydrolyse in sea water and for this reason we may expect to meet them in the latter medium either as colloids or attached to particulate matter. The other group, which does not hydrolyse as easily, will tend to occur as ions in the intestine and for this reason elements of this group will in general endanger other critical organs rather than the gastrointestinal tract, if their radioactive half life is sufficiently long. At the same time these elements will generally occur as ions in sea water. Table 4 presents a list of elements of which the chemical condition in sea water is known together with the f_1-factor selected by the I.C.R.P., which indicates the fraction of the nuclide which enters the blood from the gastrointestinal tract.

We have also listed the critical organ to be used for estimating the maximum permissible concentration for the total population. It is seen that the data in Table 4 are on the whole in very good agreement with these conclusions, although antimony seems to be out of place with 73 per cent in the ionic form, a fraction which also seems improbably high from a chemical point of view. That the gastrointestinal tract is the critical organ for molybdenum and copper is simply due to the fact that the half-life of these radioisotopes is quite short. (Any short-lived isotope has, of course, little chance to leave the gastrointestinal tract and is therefore unlikely to be concentrated in another organ.) The transition between the G.I. tract and other critical organs is in the region where f_1 has a value around 0.1. The G.I. tract is not the critical organ for pure α-emitters, because the α-particles do not reach the sensitive region of this part of the body. (For this reason the critical organ for plutonium and thorium is the skeleton. The relation between their chemical condition in sea water and the f_1-value is, of course, the same as for β-emitters.)

The elements which are easily hydrolysible and which do not occur as ions in the sea water will all suffer a very similar fate. They will be taken up by different organisms by means of a filtration mechanism and in this way enter into food chain. This gives us reason to assume that their nuclides will be taken up by different organisms in roughly the same proportion in which they occur in the sea water and that they will always retain this ratio in their passage through different animals. This assumption leads us to expect closely similar concentration factors in fish for all the nuclides which are easily hydrolysed. That this expectation agrees with observations within the accuracy of the observed concentration factors is shown in Table 5. In judging the agreement in Table 5 it should be kept in mind that the figures for ruthenium and plutonium may be on the low side. Under these circumstances the agreement does not seem to

Table 4

Element	Chemical state in sea water[6] (except when other source indicated)	Fraction leaving gastrointestinal tract (f_1)	Radioisotopes	Critical organ total population
biol. Na	ionic	1.0	Na^{22}, Na^{24}	total body
biol. K	ionic	1.0	K^{42}	total body
biol. S	ionic	1.0	S^{35}	testis
Cs	70% ionic	1.0	Cs^{134}, Cs^{135}, Cs^{137}	total body
biol. I	90% ionic	1.0	I^{131}	thyroid
biol? Mo	30% ionic	0.8	Mo^{99}	kidney, G.I.
biol. P	ionic	0.75	P^{32}	bone
biol. Ca	100% ionic[7]	0.6	Ca^{45}, Ca^{47}	bone
Sr	87% ionic	0.3	Sr^{89}, Sr^{90}	bone
Ni	90% ionic[7]	0.3	Ni^{63}	total body
biol. Cu	ionic	0.28	Cu^{64}	G.I.
Te	45% ionic	0.25	Te^{127m}, Te^{129m}	testis
biol. Zn	ionic	0.1	Zn^{65}	total body
biol. Fe	<16 ionic (84% particulate[7])	0.1	Fe^{59}	total body, G.I.
Sb	73% ionic	0.03	Sb^{124}, Sb^{125}	G.I.
Ru	0% ionic	0.03	Ru^{103}, Ru^{106}	G.I.
Ce	2% ionic	$<10^{-4}$	Ce^{141}, Ce^{144}	G.I.
Zr	1% ionic	$<10^{-4}$	Zr^{95}	G.I.
Y	0% ionic	$<10^{-4}$	Y^{90}, Y^{91}	G.I.
Nb	0% ionic	$<10^{-4}$	Nb^{95}	G.I.

The elements marked "biol." are considered to play a physiological role in the organism.

exceed the probable error of the concentration factors.

Using the fact that the concentration factors should be equal for all elements which hydrolyse readily we can make an estimate of a reasonable average concentration factor for this group to be used in formula (1). Probably the value $F_c = 8.8$ is not a bad guess. If this value is put into formula (1) we obtain for the *easily hydrolysible elements* in sea water

$$(\mathrm{MPC})_s = (\mathrm{MPC})_w \qquad (2)$$

Table 5. *Concentration factors in fish of easily hydrolysible elements*

Element	F_c
Ti	40 (very inaccurate)
Y	5
Ru	3.2
Ce	12
Pu	13

The results obtained in this formula are collected in Table 6. Evidently, most of the results given in this table will refer to the G.I. tract as the critical organ, but the formula is equally applicable to the cases of plutonium and ionium where the critical organ is the bone. Perhaps it might be better to use the observed concentrations factors for those elements where this figure is available, and we have calculated maximum permissible concentrations for sea water in this way from formula (1) and the values obtained are also found in Table 6. As was to be expected the two $(\mathrm{MPC})_s$ values differ by a factor of not more than a few units. On the whole the figures obtained from formula (2) might be the more reliable ones, however.

The equality of the concentration factors for easily hydrolysible elements should not only hold for fish but also, of course, for other organisms in the sea. This is born out by the scanty evidence available: concentration factors calculated from DUNSTER's figures[3] are thirty times higher in

Table 6. Maximum permissible concentrations for easily hydrolysible elements in sea water

Nuclide	$(MPC)_s$ taken as $=(MPC)_w$, $\mu c/ml$	$(MPC)_s$ calculated from (1) with individual F_c values from Table 3, $\mu c/ml$
Y^{91}	10^{-5}	1.8×10^{-5}
Zr^{95}	2×10^{-5}	—
Nb^{95}	3×10^{-5}	—
Ru^{103}	2.7×10^{-5}	7×10^{-5}
Ru^{106}	3×10^{-6}	8×10^{-6}
Ce^{141}	3×10^{-5}	2.2×10^{-5}
Ce^{144}	3×10^{-6}	2.2×10^{-6}
Pr^{143}	1.7×10^{-5}	—
Pm^{147}	6.7×10^{-5}	—
Sm^{151}	1.3×10^{-4}	—
Th^{230}(Io)	6.7×10^{-7}	—
Pu^{239}	1.7×10^{-6}	1.1×10^{-6}

mussels than in fish for Ru^{106} and seventy-five times higher for plutonium.

The elements for which the $(MPC)_s$ values are listed in Table 6 do not play a physiological role in the human organism as far as we know. The other elements mentioned in Table 1 on the other hand seem to have either a physiological role in the human organism or they are close chemical relations of elements which are physiologically active. (Uranium* is probably an exception as it seems to be quite unessential to life.)

PHYSIOLOGICAL ELEMENTS

Those elements which are physiologically important are in general controlled by physiological processes as far as the uptake into the body system is concerned. In other words the amount present in the body does not depend very much on the amount offered to the system. If only a small quantity is consumed most of it will be put to a good use, if much is consumed the larger part will either remain in the G.I. tract or it will be secreted by the kidneys—as a rule fairly rapidly. This means that there is a very special protection mechanism available for the human body against these radionuclides.

Sea water contains appreciable quantities of all physiological elements and once the radioactive isotope is mixed with the stable isotopes of the same element isotope separation will be impossible. In other words if the human system restricts the quantity of the element present in it, the amount of radioactivity will be limited by the *specific activity* (activity per unit weight of element) of the element in the food. If we ensure that in the sea water this specific activity does not exceed the specific activity of the element which is permissible in the food, we may be assured that the maximum permissible body burden of the radionuclide will not be exceeded, or at least will not be exceeded to a very appreciable degree. The specific activity of the element in the food can be calculated by dividing the permissible activity of the element in the daily drinking water, which is $2200 \times (MPC)_w$ (μc/day) by the daily intake of the element, I(g/day). If the concentration of the element in the sea water is expressed as c_s (g/cm^3) we shall always be fairly safe if we use the following formula:

$$(MPC)_s = c_s \cdot \frac{2200(MPC)_w}{I} \qquad (3)$$

It should be understood that this protection is, of course, only valid against over-irradiation of other organs than the intestinal tract because the uptake in the intestinal tract is not governed by the same physiological factors. This restriction will not matter much in the case of I^{131} or, to a smaller extent, Zn^{65} because there the G.I. can stand a much larger dose of the radionuclide than the critical organ. On the other hand in the case of Fe^{59}, Co^{58} and Co^{60} the quantity of the radionuclide permissible for the G.I. tract is about the same as that permissible for the other most sensitive organ and in this case formula (3) will not afford protection against over-irradiation of the G.I. tract, but it will still afford protection against radiation damage of the gonads, the blood-forming organs, etc. The maximum permissible concentrations in sea water calculated with formula (3) for isotopes of physiological elements are collected in Table 7.

Although the figures calculated in this way are

* The best $(MPC)_s$ value for uranium is probably obtained by using the F_c-value < 20 in formula (1). This gives us $(MPC)_s > 4 \times 10^{-6}$ μc/ml.

Table 7. Maximum permissible concentrations in sea water of radioactive isotopes of physiological elements

Nuclide	Critical organ	$(MPC)_w$ (μc/ml)	Daily uptake of element I (mg)	$S = \frac{2200(MPC)_w}{I}$ (mc/mg)	c_s (mg/ml)	$(MPC)_s$ from (3) (μc/ml)	F_c	$(MPC)_s$ from (1) (μc/ml)
Radionuclides which present little danger to the G.I:								
Zn^{65}	total body	10^{-5}	17	1.3×10^{-3}	10^{-5}	1.3×10^{-8}	3000	3×10^{-8}
I^{131}	thyroid	6.7×10^{-7}	0.2	7×10^{-3}	5×10^{-5}	3×10^{-7}	7	8×10^{-7}
Radionuclides which apart from other organs also endanger the G.I. tract:								
Fe^{59}	total body	2×10^{-5}	27	1.6×10^{-3}	10^{-5}	1.6×10^{-8}	1000	2×10^{-7}
	gastrointestinal tract	2×10^{-5}						2×10^{-7}
Co^{58}	total body	4×10^{-5}	0.007	13	5×10^{-7}	6×10^{-6}	160	2×10^{-6}
	gastrointestinal tract	3×10^{-5}						1.6×10^{-6}
Co^{60}	total body	10^{-5}	0.007	3	5×10^{-7}	1.6×10^{-6}	160	5×10^{-7}
	gastrointestinal tract	1.7×10^{-5}						9×10^{-7}

Table 8. Maximum permissible concentrations in sea water of radioactive isotopes of elements closely related from a chemical point of view to physiological elements

Radio-nuclide	Physiol. elements	Critical organ	$(MPC)_w$ (μc/ml)	Daily intake physiol. element I (mg)	$\frac{(F_c)\text{ physiol. element}}{(F_c)\text{ radionuclide}}$	c_s physiol. element (mg/ml)	$(MPC)_s$ from (4) (μc/ml)	(F_c) radio-nuclide	$(MPC)_s$ from (1) (μc/ml)
Sr^{89}	Ca	bone	3×10^{-6}	1000	1.0	0.4	2.6×10^{-6}	2.0	1.3×10^{-5}
Sr^{90}	Ca	bone	3×10^{-8}	1000	1.0	0.4	2.6×10^{-8}	2.0	1.3×10^{-7}
Cs^{137}	K	total body	2×10^{-6}	3000	0.4	0.4	2.3×10^{-7}	20	9×10^{-7}
Ra^{226}	Ca	total body	2×10^{-9}	1000	0.13	0.4	2×10^{-10}	15	1.2×10^{-9}

extremely safe (at least for organs other than the G.I. tract), they probably represent very serious under-estimates. If we calculate the total amount of iodine, iron and zinc taken up with 250 g of fish (for cobalt the situation is quite different) we find that this quantity of the element is appreciably less than the normal daily intake. Therefore, we may assume that a rather larger quantity of these elements enters the system with other kinds of food. This means an extra dilution of the radioactivity or, in other words, the average specific activity of the element in the food will be much lower than assumed in the calculation of formula (3). The total quantity of the element consumed per day will be I, but the total quantity of the element present in 250 g of fish will be only $F_c/8.8 \times 2200\, c_s$. Therefore, in as far as this reasoning is valid we may increase the daily intake of radioactivity and therefore the MPC in sea water by a factor equal to

$$\frac{I}{F_c/8.8 \times 2200\, c_s}$$

By introducing this factor into formula (3) in this way we again obtain a formula identical to formula (1). However, the application of formula (1) is by no means as safe as that of formula (3) because it introduces concentration factors, which in this case are not checked by their agreement with other concentration factors of elements which behave chemically in the same way. The maximum permissible concentrations for sea water obtained by the latter reasoning are also included in Table 7.

The case of the cobalt isotopes Co^{58} and Co^{60} deserves special discussion. The cobalt content of fish is so high that someone consuming 250 g of fish per day would take in much more than the normal amount of cobalt. Therefore, the $(MPC)_s$ based on total-body irradiation comes out higher from formula (3) than from formula (1). This in itself would not be a sufficient reason to prefer the lower figure since, cobalt being a physiological element, we may expect the quantity present in the body to be regulated by physiological processes. However, the fact that formula (1) applied to the irradiation of the gastrointestinal tract also restricts the $(MPC)_s$ to lower values effectively determines its value, as there is no physiological protection against over-irradiation of the G.I. tract. We therefore must accept as the $(MPC)_s$ values 1.6×10^{-6} μc/ml for Co^{58} and 9×10^{-7} μc/ml for Co^{60}.

There is another group of elements, strontium, caesium and radium which, though not isotopes of physiological elements, are very close chemical relations of physiological elements and for this reason will be very unlikely to change their concentration relative to the latter to an important degree. This is specially true in the case of strontium isotopes which have the same value for F_c as calcium, but it will also to a certain extent hold for caesium and radium, although for the latter nuclides this protection will be less certain as they are preferentially taken up by fish as indicated by the higher values of F_c for radium and caesium than for strontium and potassium. However, very important disturbances of the concentration ratio of the radionuclide and the physiological element in the sea water are unlikely to take place and therefore the physiological element will still afford very valuable protection of the human body against unexpected over-irradiation by the radionuclides. Therefore we can again apply the principle of formula (3) in these cases, but then we must use the $(MPC)_w$ value of the radionuclide in combination with the value of I for the physiological element. However, in this case we also have to take into account the fact that the ratio of the concentrations of radionuclide and physiological element will have different values in fish and in sea water. This difference is taken care of by the introduction into formula (3) of a factor $(F_c)_{\text{physiol.}}/(F_c)_{\text{radion.}}$, the ratio of the concentration factors of the physiological element and the radioelement. The formula we obtain in this way is the following

$$(MPC)_s = \frac{(F_c)_{\text{physiol.}}}{(F_c)_{\text{radion.}}} \times C_s \times \frac{2200(MPC)_w}{I} \quad (4)$$

In this way we have calculated the figures given in Table 8. In these cases too the total quantity of the physiological element consumed per day is quite a bit larger than that contained in 250 g of fish which means that an extra dilution of the physiological element takes place by a factor

$$\left[\frac{I}{(F_c)_{\text{physiol.}}/8.8 \times 2200\, C_s}\right]$$

We may therefore multiply the MPC of the radioelement in sea water by this same factor which gives us once more formula (1) in which F_c refers to the radioelement. Table 8 also shows the results obtained by this formula.

DISCUSSION

It is evident that the assumption made in calculating the MPC in sea water, i.e. that the radionuclide is distributed homogeneously throughout the sea, is by no means a realistic one. In general, very appreciable differences of concentration will be observed, the concentration being highest near a dumping place or the outlet of a pipeline. If we would make the $(MPC)_s$ values calculated in this report apply to this region of highest concentration we would err very much on the safe side. However, this will not be necessary if we take into account the following points which decrease the danger of people actually taking up the radioactivity corresponding to fish living in this area of greatest radioactivity.

(1) Some fish will move over appreciable distances and for this reason will never take up as much radioactivity as corresponds to the region of highest concentration of radionuclides. However, this is true only for some types of fish and not for others and therefore we shall not be able to introduce this diluting action into calculations. It does, however, constitute an important extra safety factor.

(2) No attention has been paid to the radioactive decay of radionuclides between the moment when they are measured in the sea water and the moment they enter into the human system from fish. For very long-lived radionuclides as Sr^{90} and Cs^{137} this is of no importance, but for other radioactive isotopes like I^{131} the radioactive decay can make a great difference. However, we have no way at present of taking this decay into account and we leave it to constitute another important factor of safety.

(3) In general the fish from the most highly contaminated region will be diluted by fish caught in other parts. To take this dilution into account it has been suggested that the allowed concentration of radioactivity in the most strongly radioactive region should be increased by a factor equal to 10.[5] In this connexion it must be kept in mind that the I.C.R.P. restrictions need be satisfied only as averages over periods of months. (Of course, such a correction factor may only be applied if the distribution of radioactivity throughout the sea is actually quite inhomogeneous. If, as the other extreme, there were an infinite number of infinitely small fishes, and mixing of the fish were complete, this would mean that our $(MPC)_s$ values could be used as values of the average concentration.)

(4) If the quantity of contaminated fish is small enough to make it certain that only a small fraction of the population can be fed with a diet containing an appreciable fraction of 250 g of fish from the radioactive region, the people reached by this food might be considered to constitute a "special group" of the type $B(c)$ according to the ICRP[3]. In this case the activity per ml of water in the region from which this fish originates may again be raised by a factor 3.

In the Appendix a simple but not very realistic model of disposing of radioactive waste into a sea is used to discuss the application of points (3) and (4) to such a process.*

No account has been taken in this report of the fact that possibly it may be decided that only part of the permissible radiation dose may be caused by fish. In that case an additional safety factor must, of course, be introduced.

Before we conclude this report it is well to consider the soundness of our line of argument. Although the distribution of the radioactivity present in the fish depends on the distribution of the concentration of the radionuclide throughout the sea, the total amount of radioactivity present in fish only depends on the average concentration in the sea of the radionuclide under consideration† which average concentration in its turn is determined by the rate of dumping and the average time a radioactive atom spends in the sea under consideration before leaving it. This means that if one were

* Of course this case does not represent conditions actually occurring. It only serves to indicate how the $(MPC)_s$ might be used under such conditions.

† It is assumed that the concentration of fish is constant throughout the sea.

only concerned with the total amount of radioactivity taken up by humans without bothering about distribution over different individuals one could determine the amount of waste to be dumped per day from the average concentration regardless of the distribution pattern. (This would, of course, be the case as we mentioned above, if there were an infinite number of infinitely small fishes and mixing of the fish were complete.) As far as genetic damage is concerned, where only the total amount of radioactivity present in the entire population plays a part, this approach to the problem would be perfectly sound. It might be considered whether the mixing of fish after the catch, the movement of fish in the sea and the changing of the fishing grounds would not make it possible to use this treatment even in connexion with the somatic damage, in which case the value of the MPC in sea water could be used without restriction as average permissible concentrations in sea water instead of as maximum values. However, under the present circumstances this would not be permissible and rules of the type discussed above will have to be followed.

APPENDIX

Application of the Maximum permissible Concentration in Sea Water to a Simple Hypothetical Case of Dumping

Let us assume that radioactive waste is being dumped into the sea at a constant rate at a fixed location. Furthermore we may assume that either the sea has a constant depth and does not show a vertical concentration gradient or that only an upper layer of constant thickness is involved in spreading the radioactivity, that this upper layer does not show a vertical concentration gradient, and that fish are taken only from this upper layer.

Next we assume that the radioactive material dumped at a certain moment spreads in such a way that it always fills a disc of water homogeneously and that the radius of this disc is proportional to time. (This assumption is roughly equivalent to the dilution process of SCHAEFER[16] except for the fact that SCHAEFER assumes the radius to be proportional to the 0.8 power of time. Although the latter assumption will certainly be more accurate, the difference is not an essential one.)

Using this assumption we find that the concentration during continuous dumping is inversely proportional to the distance between the spot where the radioactive waste is put into the sea, and the point of measurement. This means that the concentration of the radioactive nuclide (c_r) is given by (r = distance from dumping spot, Γ is a constant):

$$c_r = \frac{\Gamma}{r}$$

The concentration c_r does not vary with time.

It is immediately evident that at the place where radioactivity is dumped at the end of the pipeline the concentration of radioactive material must be very high. If we assume the fish do not move about, fish caught at this point would have a very high concentration of radioactive nuclides which would make its consumption extremely undesirable. However, this complication can easily be circumvented by forbidding all fishing activities in a certain area around the dumping spot. We shall see that this area may be quite small.

Consider a sea or a certain area which has for its boundary a circle with radius r_m, and the dumping spot at its centre. Denote the depth of the layer of water involved in the spreading of the radioactive material by h, the concentration of fish by g. The volume of this layer between the distances r and $r + \Delta r$ from the dumping spot will be $2\pi rh\Delta r$, the amount of fish contained in this volume $2\pi rhg\Delta r$. In this amount of fish we shall find an amount of radioactivity given by the formula*:

$$2\pi rhgc_rF_c\ \Delta r = 2\pi hg\Gamma F_c\Delta r$$

If we indicate the radius of the area excluded from fishing around the dumping spot by r_1, the amount of radioactivity contained in fish in the part used for fishing is given by:

$$\int_{r_1}^{r_m} 2\pi hg\Gamma F_c dr = 2\pi hg\Gamma F_c(r_m - r_1)$$

* We make the most unfavourable assumption, i.e. that the fish is in equilibrium with the radionuclide concentration on the spot.

This amount of radioactivity is contained in a quantity of fish $= \pi h g (r_m^2 - r_1^2)$. From this we find that the average concentration of radioactivity in fish

$$(c_r)_{\text{av}} = \frac{2F_c\,\Gamma}{r_m + r_1}$$

In general r_1 will be very small compared to r_m, which means that the average concentration of the radioactivity in fish will corrrespond to the equilibrium concentration half-way between the dumping spot and the outer boundary.

We now have to fit to this model our notion of the $(\text{PMC})_s$ of the radionuclide under discussion in sea water. It is, of course, tempting to put the value of $(c_r)_{\text{av}}$ equal to that of $(\text{MPC})_s$. We shall now discuss the consequences of this assumption.

The value of r_m, the radius of the sea we are discussing, must be chosen in such a way that the area covered by it may be considered to be fished as a whole and with equal intensity in its different parts so that fish in the market has been caught equally over the whole area. Although this is a fiction it probably will be a satisfactory approximation as it does not in principle tend either to raise or to lower the actual average concentration compared to the calculated value. The size of such a part of the sea must be estimated somewhat arbitrarily, but $r_m = 200$ km seems to be a reasonable choice. This means that $(c_r)_{\text{av}} = (\text{MPC})_s$ is equal to the actual value of c_r at $r = 100$ km. Of course, anywhere nearer to the dumping spot, the concentration of radioactivity in fish will be higher than $F_c \times (\text{MPC})_s$ but this does not present any complication because the higher activity in the fish from the more centrally located parts is offset by the lower activity in the fish from the more distant regions within our area, as according to the argument given in point (3), the mixing of the fish takes care of these variations. If we assume—in accordance with the British suggestion—that this averaging out may take care of concentrations of radioactivity increased up to a factor of 10, this would take care of the extra radioactivity in any fish caught at a distance of 10 km or more from the dumping spot. The amount of fish which will be caught inside a circle of a 10 km radius—if the entire area is fished with equal efficiency—will amount to only $\frac{1}{4}$ per cent of the total quantity of fish.* People who consume fish from this source may well be considered to come under the special group $B(c)$ of the ICRP and may for this reason consume fish with three times as much radioactivity as other people. This means that this extra factor makes it possible to fish anywhere outside a circle with a radius of $3\frac{1}{2}$ km from the dumping spot without exceeding the restrictions imposed by the rules of the ICRP in combination with the arguments presented in points (3) and (4).

Possibly there may be objections to this last argument, which involves the use of a special group of the population. In case this reasoning is not considered acceptable the region to be excluded will have a radius of 10 km which is still acceptable, although less satisfactory than the smaller distance.

In the preceding argument we have assumed that the radioactivity spreads equally in all directions from the dumping spot and that the latter is surrounded by water on all sides. The latter part of this assumption is by no means essential. The situation does not change much if we assume that the activity is dumped somewhere on a coast or that it is dumped at a certain spot into a stream in which it spreads laterally under a constant angle. In case the rate of spreading and the depths of the layer of water involved are known, it is a simple problem to calculate the permissible rate of dumping.

In actual cases the spreading of the activity in the water will be more complicated and a number of measurements will be necessary to obtain sufficient information concerning this process. However, this trouble will be well woth taking and after the required data have been obtained the permissible rate of dumping can be calculated.

SUMMARY

Two methods are discussed for the calculation of permissible concentrations of radionuclides in sea water, one for elements which either play a physiological role in the organism or are closely related to such elements and another method for

* Dilution by a factor 10 as discussed under (3) may bring this percentage up to 2.5.

non-physiological elements. The application of such permissible concentrations is briefly discussed.

REFERENCES

1. *Radioactive Waste Disposal into Atlantic and Gulf Coastal Waters*, National Academy of Sciences—National Research Council, P 655, Washington, D.C. (1959).
2. *Radioactive Waste Disposal From Nuclear Powerships*, National Academy of Sciences—National Research Council, P 658, Washington, D.C. (1959).
3. *Recommendations of the International Commission on Radiological Protection*, Pergamon Press, London (1959).
4. *Recommendations of the International Commission on Radiological Protection, Report of Committee II on Permissible Dose for Internal Radiation*, 1959, Pergamon Press, London (1959); *Health Phys.* **3,** (1960); *Recommendations of the National Committee on Radiation Protection, Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and in Water for Occupational Exposure*, National Bureau of Standards Handbook 69. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C.
5. H. J. Dunster, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva* Vol. 18, p. 390. United Nations, New York (1958).
6. L. Krumholz, E. D. Goldberg and H. Boroughs, In *The Biological Effects of Atomic Radiation* p. 69. Publication No. 551, National Academy of Sciences—National Research Council, Washington (1957).
7. *The Effects of Atomic Radiation with regard to the Specific Questions concerned with the Oceanography and Marine Biology, in respect to the Disposal of Radioactive Wastes*, FAO.57/7/4725, Food and Agriculture Organization (Fisheries Division) and World Health Organization.
8. F. A. Richards, *Physics and Chemistry of the Earth* Vol. 2, p. 77. Pergamon Press, London (1957).
9. D. P. Malinga. Quoted in A. P. Vinogradov, *Vernadski Laboratory for Geochemical Problems*. Memoir II of the Sears Foundation of Marine Research, New Haven (1953).
10. H. J. Dunster, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1955 Vol. 9, p. 712 United Nations, New York (1956).
11. H. Boroughs, W. A. Chipman and Th. Rice, *The Biological Effects of Atomic Radiation* p. 80. Publication 551, National Academy of Sciences—National Research Council, Washington, D.C. (1957).
12. W. A. Chipman, *Accumulation of Radioactive Materials by Fishery Organisms*. Paper presented at the 11th Annual Meeting of the Gulf and Caribbean Fisheries Institute, Miami Beach, Florida, 17–21 November (1958).
13. P. O. Agnedal, N. E. Barring, J. S. Lindke and J. W. Smith, *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva* Vol. 18, p. 400. United Nations, New York (1958).
14. W. V. Mayneord, J. M. Radley and R. C. Turner, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva* Vol. 23, p. 150. United Nations, New York (1958).
15. F. F. Koczy, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva* Vol. 18, p. 351 United Nations, New York (1958).
16. M. B. Schaefer, In *The Biological Effects of Atomic Radiation* p. 133. Publication No. 551, National Academy of Sciences—National Research Council, Washington (1957).

Health Physics Pergamon Press 1961. Vol. 6, pp. 126–135. Printed in Northern Ireland

RADIATION EFFECTS OF A 10-MW REACTOR ON ENVIRONS OF THE GEORGIA NUCLEAR LABORATORIES*

J. M. SELBY†, C. A. WILLIS, B. M. BOWEN and J. H. EDGERTON
Nuclear Safety Department, Lockheed Nuclear Products, Lockheed Aircraft Corporation, Georgia Division, Marietta, Georgia

(*Received* 15 *November* 1960)

Abstract—The Radiation Effects Reactor (RER) has been operated for approximately 600 MWh. During that time, studies have been made to determine the radiation and activation effects on the environs of the Georgia Nuclear Laboratories (AFP 67). Soil, air and water samples have been collected to determine the amount of activation and the resulting buildup of radioactive materials. Instrument surveys and foil measurements have been made to determine radiation levels. The effects of high γ- and neutron exposures on plant life have been observed as part of an ecology program performed in conjunction with Emory University. This paper is a preliminary report of these studies.

INTRODUCTION

THE main reactor facility at the Georgia Nuclear Laboratories (GNL), operated by Lockheed Nuclear Products for the United States Air Force, is a 10-MW reactor built for use in providing nuclear aircraft design data. In order to provide a large irradiation area capable of handling aircraft systems, this reactor was designed to operate in the open, utilizing air shielding and distance for the protection of personnel. This paper is a description of the effects of radiation on the environment.

The location of the 10,000 acre site, approximately 50 miles north of Atlanta in a sparsely populated area in the foothills of the Blue Ridge Mountains, is shown in Fig. 1.[1] The Georgia Nuclear Laboratories consist of the Radiation Effects Reactor (RER) and Critical Experiment Reactor (CER) designed by the Atomic Power Equipment Department of General Electric Company, San Jose, California, from a conceptual design by the Nuclear Branch of the Georgia Division of the Lockheed Aircraft Corporation, Marietta, Georgia; Radiation Effects Laboratory (REL) and Nuclear Support Laboratories. The Critical Experiment Reactor (CER) is used to determine core configurations for the Radiation Effects Reactor (RER), where the components and subsystems are irradiated. Irradiated materials are transferred from the RER to the hot cells in the Radiation Effects Laboratory (REL) by means of the railroad transfer system shown in Fig. 2.

The Radiation Effects Reactor, shown in Fig. 3, is a heterogeneous, air shielded, pressurized light water moderated and cooled reactor.[2] It is designed to operate at 10 MW with a high fast neutron leakage current of 2.8×10^{11}. The core consists of twenty-six modified MTR type fuel elements, four fuel cadmium poison control rods, and a cadmium-silver regulating rod. The core contains approximately 5.4 kg of uranium highly enriched in U^{235}. It is contained in an aluminum inner tank, a stainless steel pressure vessel, and an aluminum shield tank containing variable water thicknesses. The shield tank is encircled by a boral shield with a removable section at the northeast face. The reactor is supported by a hydraulic lift, which raises above the 38-ft deep pool for operation. The reactor is remotely operated from an underground control room in a concrete building.

Operation performed at a reactor site creates many radiation control and safety problems. One is the release of solid, liquid, and gaseous

* Air force plant no. 67.

† Present Address; General Electric Company, Hanford Atomic Products Operation, Richland, Washington

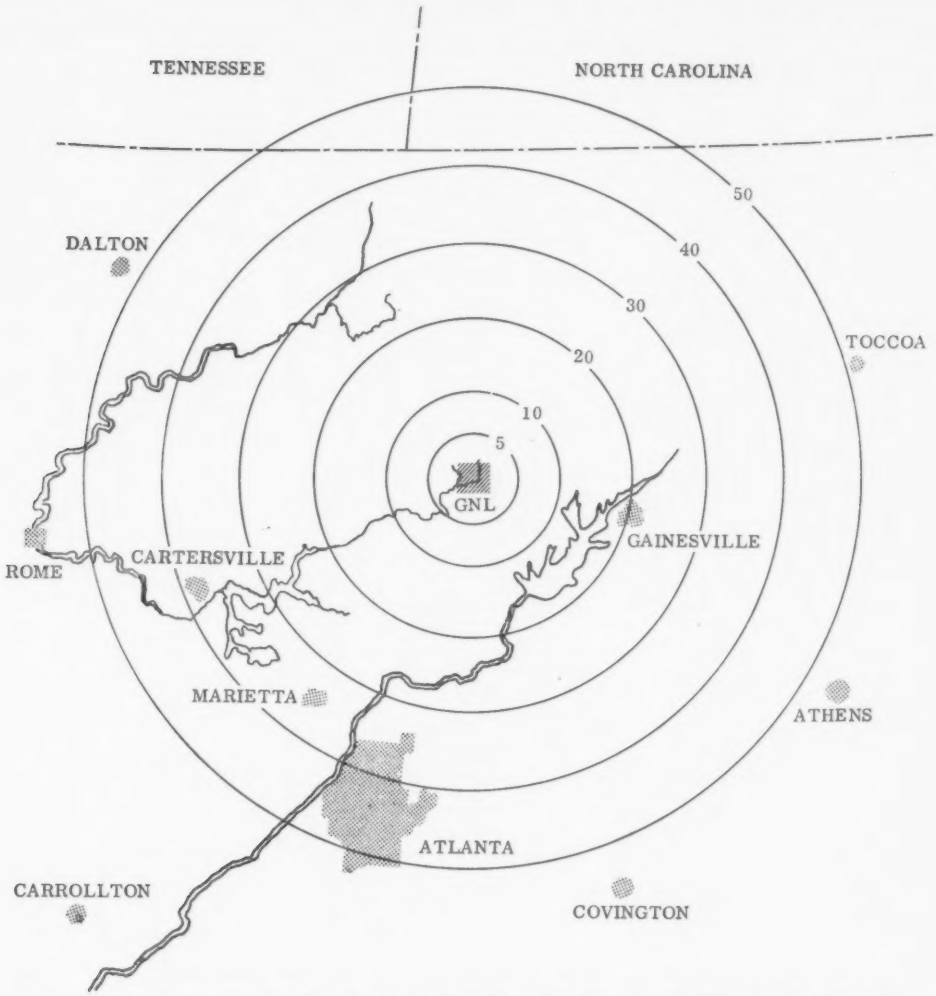


FIG. 1. Cities and counties at 5-10-20-30-40-50 mile radius from Georgia Nuclear Laboratories.

radioactive materials and their effects on the environment.[3] Release of radioactive materials to the environs may occur in the following ways:

(1) Emission of small undetected amounts of radioactive materials over a long period of time.
(2) Controlled discharge of radioactive waste to the environs.
(3) Direct activation of materials around reactor building with subsequent erosion and release to the environs.

The radioactive materials released to the environs may eventually enter a human body by inhalation, ingestion, or by transfer through the food chain. Because of toxic effects or radiation damage resulting from internal deposition of radioactive materials, the National Committee for Radiation Protection has established rigid limits (maximum permissible concentrations) for the radioactivity density in water and air.

To ensure that operation of the air-shielded reactor will create no problem to persons living

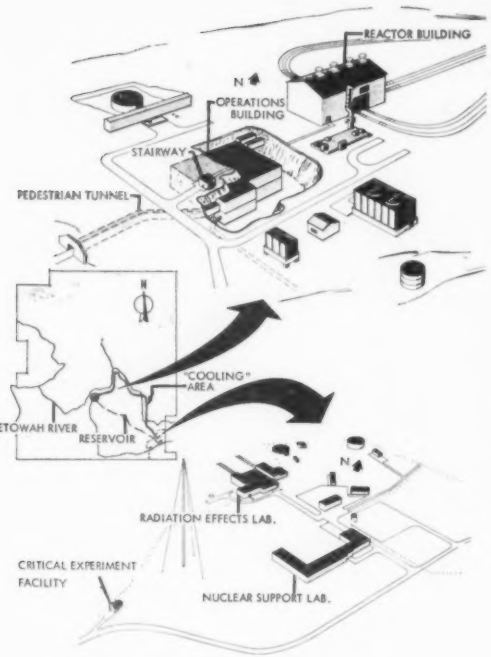


FIG. 2. Georgia Nuclear Laboratories.

near the site, it was necessary to determine the buildup of radioactive materials in soil, air, and water and to observe the effects on the plant ecology with reference to possible future erosion problems.

DUST ACTIVATION

As shown in Fig. 4, air samplers were located at three positions near the reactor during the 100 hr of continuous 1-MW operation and at seven positions near the reactor during 10-MW operation. The samplers were operated continuously before, during, and after the 10-MW operation. The air filters were counted for decay in a 7-in. proportional flow counter equipped with an end window. During one period of 10-MW operation under inversion conditions, a large volume sample was collected with an electrostatic precipitator at the Nuclear Support Laboratory and counted in a NaI deep-well scintillation spectrophotometer for decay.

During 1-MW operation, there was no detectable increase in activity over concentrations found before operation. During the 10-MW operation, however, the concentration between 250 ft and 80 ft increased by a factor of approximately 200, as illustrated in

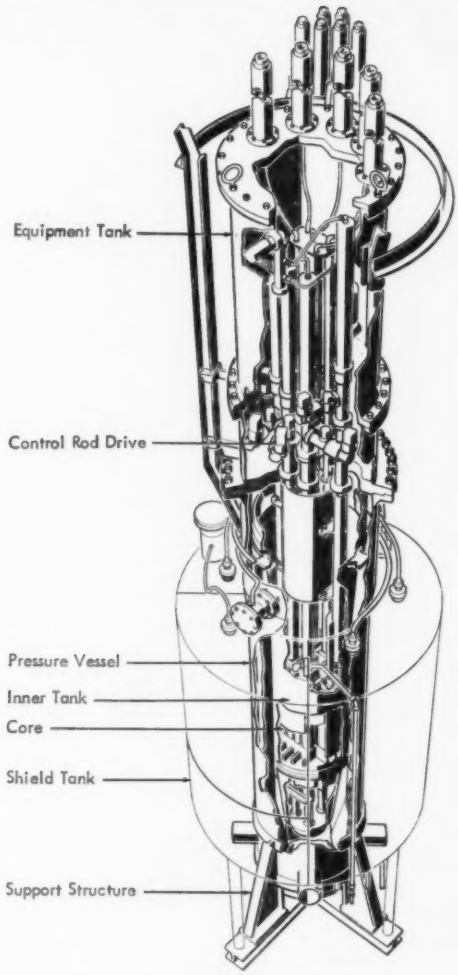


FIG. 3. Radiation effects reactor.

Fig. 5. The concentrations at greater distances during 10 MW operation varied between 1×10^{-11} and 3×10^{-11} μc/ml, which may be related to changes in meteorological conditions and not to reactor operations.

Analyses of the decay characteristics of the air samples indicates an effective 5-hr half-life during 10-MW operation, which is similar to the soil decay shown in Fig. 11. The activity in the 10-MW air samples can therefore be related primarily to Mn^{56} ($T_{1/2} = 2.68$ hr) and Na^{24} ($T_{1/2} = 15$ hr). All air samples at 1 MW and samples collected at distances greater than 250 ft at 10 MW exhibited decays

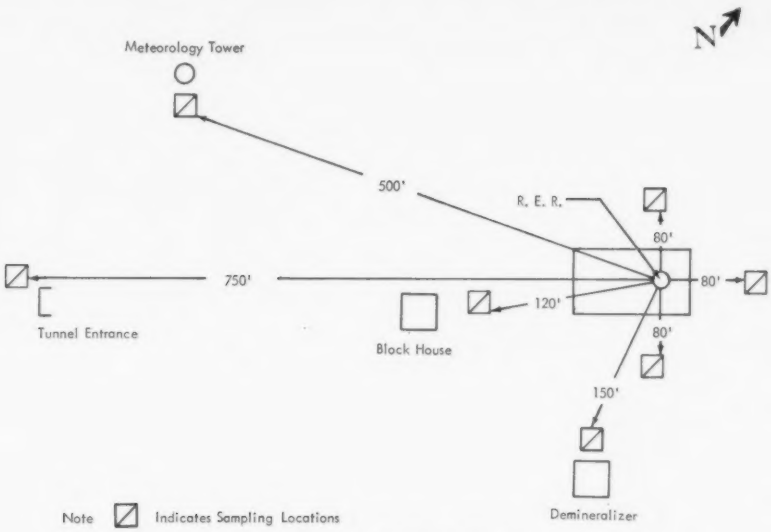


FIG. 4. Dust sample location.

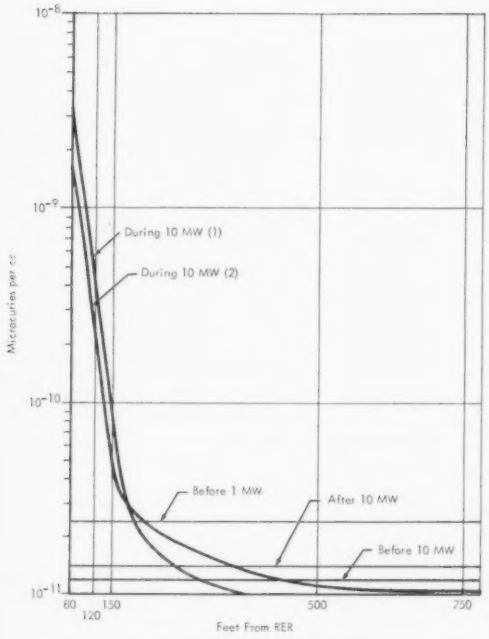


FIG. 5. Dust activation vs. distance

similar to those associated with naturally occurring isotopes. The air sample collected at the NSL during the 10-MW operation also exhibited the characteristic decay curve associated with naturally occurring isotopes.

The radioactivity density in air after shutdown is sufficient to require the use of masks in the vicinity of the reactor. However, at distances greater than a few hundred feet, because of air shielding and meteorological dilution factors, radioactive concentrations in air present no problem.

ECOLOGICAL OBSERVATIONS

The biological effects of radiation are primarily of interest to biologists; however, because of the relation of plants to the food chain and the possible increase of soil erosion from an area affected by radiation, any adverse effects from reactor operation will be of concern to radiation protection personnel. Emory University, as one part of its study of the long-range effects of radiation on the environment, has been closely observing the plant life during the initial stages of operation. Although it is too early to make any definite conclusions about the plant life around the reactor, certain trends which could be predicted from past experimental work are apparent.

Pine trees, which are known to be radiation

sensitive, are brown and appear to be dead within 500 ft of the reactor.[4] Pine trees in the area from 500 to 700 ft from the reactor show mixed results, while most pines outside 700 ft are still green and exhibit new growth. Within 300 ft of the reactor where the soil is sandy, crab grass (*Digitaria*) is increasing where vegetation is not present in thick cover. Crab grass is very resistant to radiation damage, as reported by CARTER.[5] Crab grass is reported to resist radiation in chronic dose up to 6000 r per day. It is difficult, from the limited amount of operation to date, to make any long-term predictions. It is logical to assume, however, that radiation-resistant plants, such as crab grass, will replace radiation sensitive plants in the affected areas.

FLUX MAPPING

The activation of manganese was chosen as one means of determining the neutron distribution in the 3600-ft enclosed area around the RER. Manganese dioxide and pure manganese metal were exposed in polyethylene tubes in weights equivalent to 4 or 5 g of the pure element. The weight of each specimen was recorded to the nearest milligram.

During 10-MW operation, the specimens were placed at various locations around the reactor and out to 3000 ft. The tubes were placed in 1-in. thick polyethylene moderators to make exposure conditions uniform. During one irradiation period, the reactor shield tanks were full and the boral shield was on; during the other irradiation period, the tanks were empty and the boral shield was removed. These two extremes were used to provide data on the maximum and minimum flux expected during 10-MW operation.

The specimens were counted on a NaI deep-well scintillation crystal connected to a single-channel analyzer. Using the decay rate of the pure element, the reactor power, and amount of operation, the data were normalized back to the time of reactor shutdown.

The data from the two extremes were plotted on Fig. 6 in n/cm^2 per sec at varying distances up to 3600 ft.[6] The results from the two operations compare favorably and they vary by a factor of approximately 200. If an average

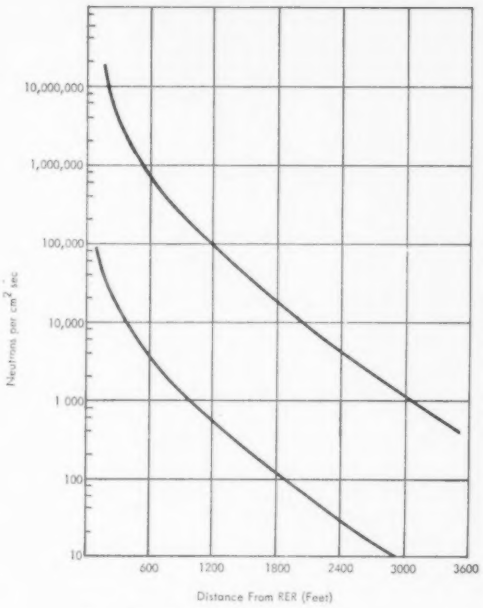


FIG. 6. Neutron flux vs. distance.

RBE of 10 is used as suggested in NBS handbook 63, where 4 n/cm^2 per sec is equivalent to 1 mrem/hr. the dose rate under maximum conditions is 100 mrem/hr at the reactor exclusion fence.

Instrument surveys were made within the 3600 ft area during 1-MW and 10-MW operation with tanks empty and boral shield off to supplement the neutron foil measurements and γ- and neutron film measurements. As should be expected, measurements at some locations vary by a factor of approximately 10, depending on the power level. Measurements were also made to determine the difference in dose rate when the tanks were full and the boral shield was on. The neutron dose rate dropped by a factor of 50 and the γ-dose rate dropped by a factor of 4. The difference between the factor of 50 with a neutron instrument and factor of 200 with the foils probably results from the fact that the foils were in line of sight of the reactor, while all instrument readings were taken along existing roads, which are terrain shielded. The instrument reading at the 3600 ft fence in an unshielded location compared

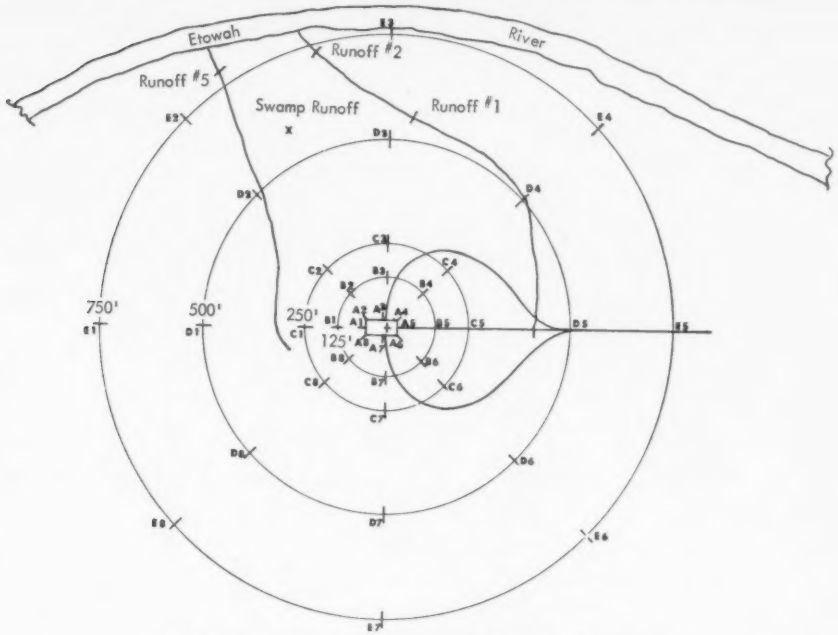


FIG. 7. Soil and runoff water sample collection locations.

favorably with the neutron flux measurement at the same location.

SOIL ACTIVATION

The radioactivity induced in the soil by escape neutrons from the RER is evaluated by regularly analyzing soil samples. These samples are taken from the locations indicated in Fig. 7. Other samples are analyzed from locations as far from the RER as the exclusion fence, 3600 ft away. The samples are counted for gross β-radiation using an end window proportional counter.

The gross β–γ activation vs. distance relationship is shown in Fig. 8; the curve has been smoothed somewhat to eliminate the effects of terrain variation. Comparing this with Fig. 6 shows that the activity concentrations fall off more rapidly with distance than does the neutron flux. This is assumed to be the result of the increase in mean neutron energy with distance. The curve falls off less sharply than the inverse square relationship indicates, out to a distance of about 250 ft. This is assumed to be the result of the thermalization of intermediate energy neutrons.

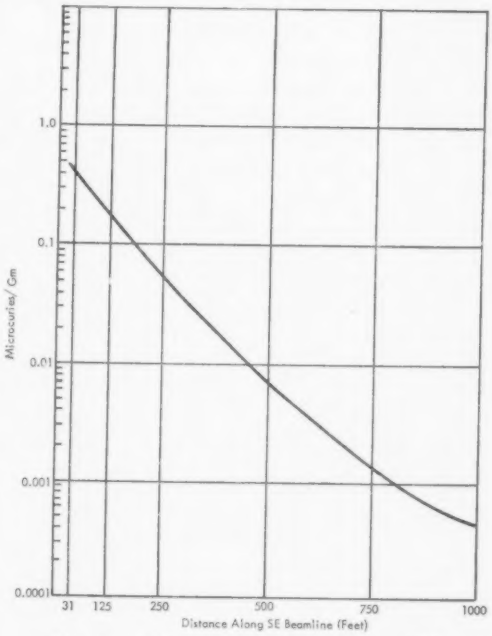


FIG. 8. Activation vs. distance from RER after 21 hr at 10 MW.

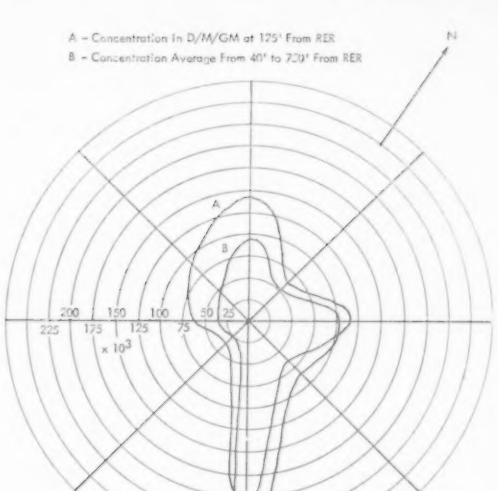


FIG. 9. The angular distribution of soil activation after 10 MW operation.

The radioactive material in the soil is not distributed symmetrically about the RER. The effects of the boral shield, buildings, and terrain variations combine with the flux distribution to give the maximum concentrations along the southeast and northwest beam lines. Fig. 9 shows the activity–angle relationship.

The soil activity density decreases with increase in depth. Fig. 10 illustrates this relationship at 125 ft from the reactor in the southeast beamline. In a box of soil placed 3 ft from the RER, the thermal neutron flux was observed to be slightly greater at a depth of 3 in. than at the surface. The experiment was not sufficiently refined to allow observation of the corresponding variation in activation. At distances of interest, (>30 ft) this effect is not observed; the maximum activation occurs at the surface.

The induced activity in the soil is, of course, primarily from radioisotopes with short half lives. An isotope with a half life greater than 100 days would not be present in a concentration greater than 1 per cent of its equilibrium value. Fig. 11 shows the typical decay characteristics; these two samples were collected immediately after 10-MW operation. For the first few hours, samples showed an effective 5-hr half life. This probably results from the decay of (2.58 hr) Mn^{56} and (15 hr) Na^{24} in combination. Both isotopes have been identified from γ-spectroanalysis. The effective half life then increases to about 18 hr, probably as a result of Na^{24} mixing with several slightly longer lived unidentified radioisotopes. The long half life exhibited after several days decay is probably due to naturally occurring radioisotopes.

There has been little buildup of activity in the soil from the 1-MW and 10-MW operation. Samples were collected and analyzed before criticality, before 1-MW operation, before the 100-hr 1-MW operational period, after the 100-hr operation, and after the 10-MW operation. Table 1 gives the average concentrations at each of the regular sample distances shown in Fig. 7 and at the exclusion fence. It shows that, after a 7-day decay period, the soil activity is little greater beyond 500 ft than it was before irradiation. The slight increase shown is less than might be expected from the high fallout concentrations observed in rain water over this

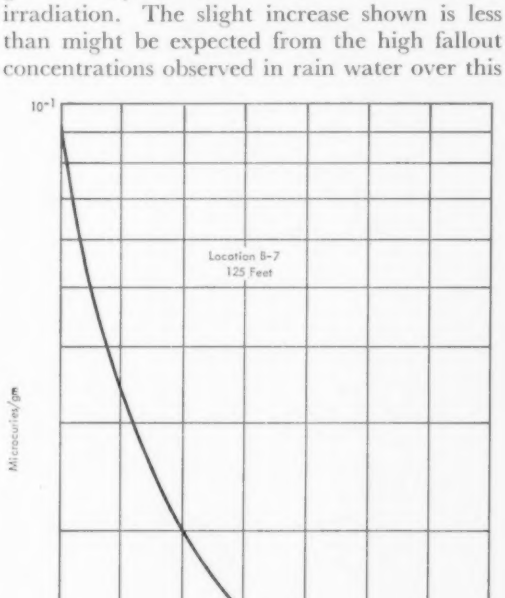


FIG. 10. Soil activation vs. depth.

period. The activity in the soil near the RER is still high compared to background, but it is still decaying with a short effective half life.

Investigations are being conducted to identify the radioisotopic constituents of the irradiated soil. Soil samples are subjected to γ-spectro-analysis, β-energy determinations, and decay characteristic investigations. Special samples have been irradiated within 3 ft of the RER and subsequently used in isotope determinations. Samples of the soil from the RER area were irradiated at Oak Ridge National Laboratory; these were investigated for isotopic content at Oak Ridge. Many radioisotopes have been detected, and others are suspected. The failure of some isotopes of interest (P^{32}, S^{35}, and Ca^{45}) to emit γ-radiation complicates their identification.

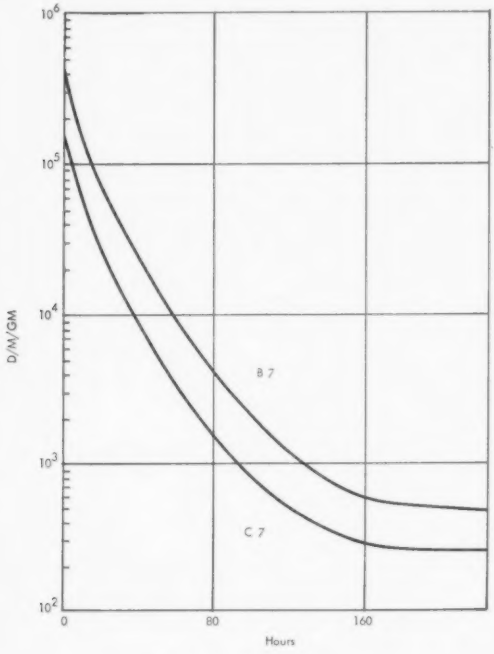


FIG. 11. Soil decay.

Of the thirty plus radioisotopes that have been at least tentatively identified, only a few are expected to present a health physics problem. These are listed in Table 2. The soil columns show the maximum concentrations expected in the soil based on 10^7 nV and chemical analyses of the soil. Where more than one nuclear reaction results in the radionuclide of interest, all have been considered; for example, $Mn^{55}(n, \gamma)Mn^{56}$ and $Fe^{56}(n, p)Mn^{56}$. In most instances the neutron–γ reaction predominates. There has been enough experimental evidence to show that high-energy reactions contribute only slightly. Radioisotopes not included here are not expected to appear in significant quantities in the soil, even after many years of reactor operations. The max. column lists the equilibrium concentration. The 100-hr column

Table 1. *Average β–γ concentration in soil around RER*
($\mu c/g \times 10^{-3}$)

Ring	I	II	III	IV	V	VI
A 50 ft (ave.)	0.054	0.068	0.13	0.34	132	2.5
B 125 ft	0.043	0.051	0.12	0.15	44	0.56
C 250 ft	0.069	0.087	0.13	0.11	15	0.34
D 500 ft	0.068	0.068	0.13	0.09	2.2	0.16
E 750 ft	0.069	0.060	0.09	0.07	1.1	0.10
Fence 3600 ft	No sample	No sample	0.13	0.08	0.08*	0.10

I. Collected December 1958 (before first RER criticality)
II. Collected 17 March 1959 (prior to 1-MW operation)
III. Collected 3 June 1959 (prior to 100 hr continuous 1-MW operation)
IV. Collected 12 June 1959 (counted 16 June after 1-MW operation)
V. Collected 20 June 1959 (counted 24 hr after shutdown 10-MW operation)
VI. Collected 20 June 1959 (counted on 26 June 1959—7 days after 10-MW operation)

* Counted 4 hr after shutdown.

Table 2. Soil radioisotopes

Radioisotope	$T^{1/2}$	MPC	Soil†			Max. runoff‡	
			Present (%)	100 hr	Max.	Soluble	Insoluble
Na^{24}	15 hr	8×10^{-4}	45	4.6×10^{-2}	5.5×10^{-2}	10^{-5}	2×10^{-5}
Si^{31}	2.6 hr	6×10^{-3}	95	4.6×10^{-3}	4.7×10^{-3}	2.1×10^{-7}	1.9×10^{-5}
*P^{32}	14.5 days	5×10^{-4}	7.5	2.2×10^{-3}	5.4×10^{-3}	10^{-7}	1.5×10^{-5}
*S^{35}(?)	87 days	2×10^{-3}	0.8	3.0×10^{-5}	1.3×10^{-3}	3.8×10^{-7}	10^{-6}
K^{42}	12.5 hr	6×10^{-4}	46	2.5×10^{-3}	4.4×10^{-3}	5.4×10^{-7}	4.3×10^{-6}
*Ca^{45}(?)	160 days	3×10^{-4}	0.6	6.5×10^{-5}	4.6×10^{-3}	8.8×10^{-7}	1.4×10^{-5}
Mn^{56}	2.58 hr	3×10^{-3}	95	4.9×10^{-2}	5.1×10^{-2}	—	—
Fe^{59}	45 days	2×10^{-3}	3	4.5×10^{-5}	2.3×10^{-3}	2.4×10^{-7}	2.4×10^{-5}
As^{76}	26.7 hr	6×10^{-4}	44	4.0×10^{-4}	9.3×10^{-4}	—	—

* No γ.
† $\mu c/g$.
‡ $\mu c/cm^3$.
(?) Only tentative identification.
MPC occupation MPC for water in $\mu c/cm^3$.
$T_{1/2}$, half-life.

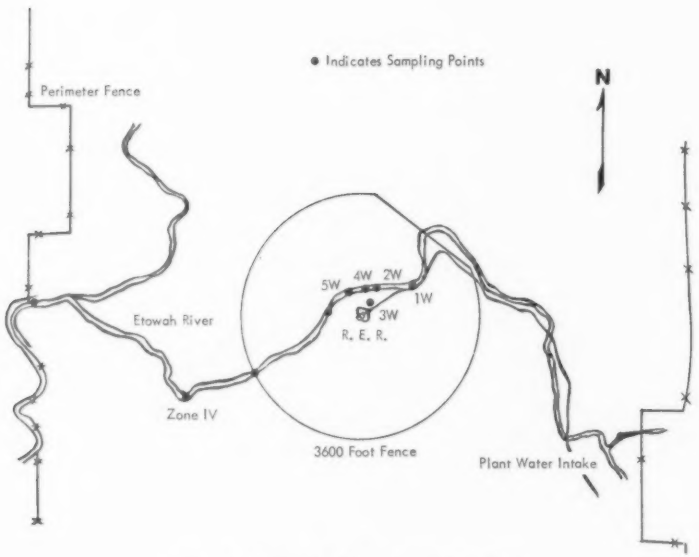


FIG. 12. Etowah River sampling sites.

lists the concentration expected after 100 hr of intermittent 10-MW reactor operation. The present column lists the fraction of the equilibrium value achieved in the recent 42 hr of intermittent 10-MW reactor operation. The max. runoff columns show the maximum concentration to be expected in the runoff water before dilution by the Etowah River. These values are based on chemical analysis of runoff water and equilibrium activation by 10^7 nV. The maximum observed concentration of the element was used in each case.

The soil activation study is not complete. Sufficient information has been obtained to establish that the hazard from activated material washing into the Etowah River is significant but,

at this time, not excessive. Further soil activation studies are being conducted in conjunction with, and to augment, a soil stabilization study.

WATER ACTIVATION

Water samples are collected weekly from several locations shown in Fig. 12 above and below the reactor site. Five hundred milliliters from each weekly grab sample are concentrated by drying and are counted for gross β–γ activity in a proportional flow counter equipped with an end window.

Continuous samples are collected by Georgia Public Health Department from locations above and below AFP 67. Duplicate samples are supplied weekly to Lockheed.

Continuous samplers and grab samplers are located on two runoff streams where water flows during periods of precipitation. These two streams drain approximately 95 per cent of the activated area around the reactor.

Data from weekly grab samples collected during the period of study are tabulated in Table 3. There is considerable fluctuation in radioactivity density with the highest average concentration occurring after 1-MW operation. This fluctuation does not result from reactor operation, but is rather a normal fluctuation in concentration of radioactive materials in river water.

Table 3. *Radioactivity in water samples*

Etowah River grab samples (μc/ml $\times 10^{-9}$)

Location	I	II	III
Plant water intake	2.9	3.2	1.3
1 W	1.6	1.5	1.0
2 W	0.58	0.95	2.1
3 W	0.49	2.8	2.8
4 W	2.9	2.8	0.32
5 W	3.8	1.9	0.14
3600-ft fence	0.19	1.0	0.31
Zone IV	2.6	2.2	1.0
Perimeter fence	0.52	1.5	3.9
Average	1.7	2.0	1.6

I. Prior to 100-hr continuous 1-MW operation
II. After 100-hr continuous 1-MW operation
III. After 10-MW operation

Approximately 0.8 in. of rain fell in $\frac{3}{4}$ hr during one period of 10-MW operation. Runoff samples were collected during the rain and returned to the laboratory for processing at the end of the shift. The highest radioactive density was 1.2×10^{-6} μc/ml, with an average for the four samples of 6.8×10^{-7} μc/ml, The runoff water contained approximately 1 g of sediment in 500 ml, compared to approximately 0.01 g in river water. Water collected from the Georgia Public Health during the period in which the rain fell and the reactor was operating contained 1.3×10^{-9} μc/ml.

Based on Na^{24}, the principal activity found in irradiated soil, the maximum permissible concentration (MPC) as listed in NBS handbook 69 for offsite personnel is 3×10^{-5} μc/ml. Even before dilution, conservatively estimated at a factor 100, the maximum found in runoff water was a factor of 25 lower than the offsite MPC.

Acknowledgments—The assistance of J. E. Bramblett, K. R. Holtzinger, J. A. Mohrbacher, W. T. Price, Newton Underwood (North Carolina State College Consultant), N. W. White, J. R. Wright, and members of the Nuclear Measurement Department Radiochemistry Laboratory in the preparation of this paper is gratefully acknowledged.

REFERENCES

1. *Radiation Effects Reactor Safeguards*, LAC 147, 15 January (1958).
2. J. H. Edgerton, *Analytical Requirements of the Lockheed Critical Experiment Reactor, of the Radiation Effects Reactor, and of the Radioactive Waste Disposal System.* AEC TID 7568, Pt. 1, pp. 50–74, February (1959).
3. J. M. Selby and R. E. Simpson, *A Determination of the Concentrations of Radioactive Material at AFP* 67. LAC NR-52, July (1959).
4. C. P. Daniel, J. T. McGinnis and R. A. Pedigo, *Ecological Observations.* Unpublished Report, Emory University Ecological Group, July (1959).
5. T. C. Carter, *Biological Sciences, Progress in Nuclear Energy* Vol. 6. Pergamon Press, London (1956).
6. N. Underwood, Calculations of Neutron Flux, North Carolina State College Consultant.

Health Physics Pergamon Press 1961. Vol. 6, pp. 136–141. Printed in Northern Ireland

EVALUATION OF THE VENTILATION RATE FROM THE DECAY OF FILTER-PAPER AIR SAMPLES OF THORON OR RADON DAUGHTERS*

K. S. SOMAYAJI
Atomic Energy Establishment Trombay, Bombay, India

(*Received* 11 *July* 1960)

Abstract—The inhalation hazard associated with the processing of thorium and uranium is mostly due to the daughter products of thoron and radon than to the parent gases themselves. With the provision of ventilation the equilibrium between the parent and the decay products is disturbed resulting in a marked reduction of the inhalation hazard. TSIVOGLOU *et al.* described a method of estimating the relative proportions of the various daughter products of radon in the atmosphere, based on an analysis of the shape of the α-decay curves of filter-paper air samples.

This paper describes a simple method of obtaining the ventilation rate in a thoron field and the relative non-equilibrium among the decay products of thoron. It is indicated how the ratio of the α-activity after a delay of 150 min to that after a delay of 30 min can be used as a convenient parameter to do this.

Curves are given showing the variation of the potential α-energy values for thoron and radon daughters with the ventilation rate, which are useful when planning control by means of ventilation.

INTRODUCTION

THE major health hazards associated with the processing of uranium and thorium arise out of the continuous generation of radon and thoron. Being gases, they diffuse profusely into the atmosphere and soon decay, owing to their short half-lives, into their daughter products which attach themselves to the numerous submicron particles always present there, thus giving rise to what are called radioactive aerosols. The hazards thus arise not only out of the direct inhalation of radon and thoron but also out of the inhalation of these aerosols. Moreover, directly inhaled radon or thoron is largely breathed out before it has appreciably decayed while a significant fraction of the inhaled aerosols is retained in the lungs. The dose delivered to the lungs by the decay products of radon and thoron is, therefore, much higher than that delivered by the parent gases themselves.[1–5]

Adequate ventilation in a uranium mine is known to have reduced this hazard markedly inasmuch as it lowers the radon level in the mine as well as the equilibrium concentrations of the daughter products.[6–9] TSIVOGLOU *et al.*[10] described a method of estimating these equilibrium concentrations by taking filter-paper air samples and studying their decay. The shape of the α-decay curves of these samples shows significant variations from their shape under no-ventilation conditions during the first 45 min after sampling and yields, when analysed, the relative proportions of the various daughter products in the atmosphere.

Since the equilibrium concentrations are largely governed by the rate of ventilation in the mine, it should be possible to estimate the ventilation rate obtaining in the mine directly from the shape of the decay curves. The present paper gives a simple and convenient method of doing this in the case of a thoron field. It is of interest inasmuch as it enables a correct evaluation of the breathing hazard in thoron fields and hence a satisfactory approach

* Paper presented on 1 July 1960 at the Applied Health Physics Session, 1960 Annual Meeting of the Health Physics Society, held at Boston, Massachusetts.

Table 1. *Nuclear properties of thoron and daughters*

Isotope	Half-life	Radiation	Energy (MeV)
Rn^{220} (Tn)	54.5 sec	α	6.28
Po^{216} (ThA)	0.158 sec	α	6.77
Pb^{212} (ThB)	10.6 hr	β	0.36
Bi^{212} (ThC)	60.5 min	66.3% β	2.25
		33.7% α	6.05
Po^{212} (ThC′)	3.04×10^{-7} sec	α	8.78
Tl^{208} (ThC″)	3.1 min	β	1.79
Pb^{208} (ThD)	Stable		

to the ventilation requirements therein. This method will be illustrated with a worked example towards the end of the paper. In principle, it can be applied to radon atmospheres as well. The case of radon, however, is not dealt with here as the effect is not very well pronounced. The relevant decay characteristics of the thoron series are given in Table 1.

PRINCIPLE OF THE METHOD

Consider a room in which thoron is generated at a constant rate. Let us assume that the ventilation is such that p parts of the air in the room are exchanged per minute at a uniform and constant rate, for fresh air which is practically free from thoron and which mixes with the room air almost instantaneously. Under no-ventilation conditions, the equilibrium concentrations of thoron and its daughter products in the room will depend only on the rate of generation of thoron and the radioactive decay constants of these elements. As soon as the ventilating system is started, however, the equilibrium will be disturbed and the concentrations will decrease steadily and finally attain new stationary values, considerably lower than the no-ventilation values. Under the new steady-state conditions, let the concentrations of ThA, ThB, ThC, ThC′ and ThC″ in the room be respectively f_1, f_2, f_3, f_4 and f_5 times their respective equilibrium concentrations. Let λ_1, λ_2, λ_3, λ_4 and λ_5 be the respective radioactive decay constants in min^{-1} units. Then, as shown by HULTQVIST[11]

$$f_1^{-1} = (1 + p/\lambda_1)$$
$$f_2^{-1} = f_1^{-1}(1 + p/\lambda_2)$$
$$f_3^{-1} = f_2^{-1}(1 + p/\lambda_3)$$
$$f_4^{-1} = f_3^{-1}(1 + p/\lambda_4)$$

and

$$f_5^{-1} = f_3^{-1}(1 + p/\lambda_5)$$

Table 2. α- *and* β-*activities of* 1 hr *filter paper-samples for different rates of ventilation**

p (min^{-1})	f_2	α-Activity		β-Activity	
		(dis/min)	Normalized	(dis/min)	Normalized
0	1.	44.93	1.	87.86	1.
10^{-5}	0.9909	44.51	0.9905	87.04	0.9907
10^{-4}	0.9160	40.97	0.9118	80.29	0.9138
10^{-3}	0.5215	22.45	0.4996	44.82	0.5101
10^{-2}	9.827×10^{-2}	3.340	7.434×10^{-2}	7.539	8.581×10^{-2}
10^{-1}	1.078×10^{-2}	2.573×10^{-1}	5.726×10^{-3}	7.165×10^{-1}	8.155×10^{-3}
10^{0}	1.084×10^{-3}	2.364×10^{-2}	5.262×10^{-4}	6.996×10^{-2}	7.963×10^{-4}
10^{1}	1.050×10^{-4}	2.341×10^{-3}	5.209×10^{-5}	6.976×10^{-3}	7.940×10^{-5}

* Concentration of thoron = 1 atom/l.
Suction rate = 1 l/min.
Delay time = 30 min.

The thoron concentration is reduced by a factor $(1 + p/\lambda_0)$ from that under no-ventilation conditions, λ_0 being the decay constant of thoron in min^{-1} units.

If now the air in the room is passed through a filter-paper (assumed filtration efficiency 100 per cent) by means of a suction pump, the decay products which are attached to dust particles, are retained on it. If we assume the suction rate to be small so that the general distribution of the air contamination is not appreciably altered, the total number of atoms of the different daughter products on the filter-paper at the end of a sampling time, t, can be calculated using the above equations. Knowing these numbers, the α- and β-activities of the filter-paper sample at the end of any interval of time, T, after the sampling has been completed can be estimated by taking into consideration the disintegration scheme and the decay constants of the various elements present on the filter-paper. Some of the values thus obtained are given in Table 2 and correspond to the case when the collection time $t = 60$ min and the delay time $T = 30$ min. For the purpose of these calculations, the number of thoron atoms per litre of air under the steady state is assumed to be unity. These values are normalized by taking the no-ventilation value of the activity as unity. The values of f_2 are also given in the table. It is seen that the normalized activity of the sample for a fixed steady-state concentration of thoron varies in the same manner as the value of f_2, indicating that the air contamination is mainly due to ThB, which is true.

The activities of the samples corresponding to the values of T varying up to 300 min have been computed and normalized taking the value at $T = 30$ min (A_{30}) as unity. The manner in which the activity of the sample changes with time depends upon the rate of ventilation. The normalized α- and β-decay curves for different values of p are shown in Fig. 1 and Fig. 2, respectively. It is clear that the shape of the decay curves is remarkedly sensitive to variations of p in the range from 10^{-3} to about 1, which one normally comes

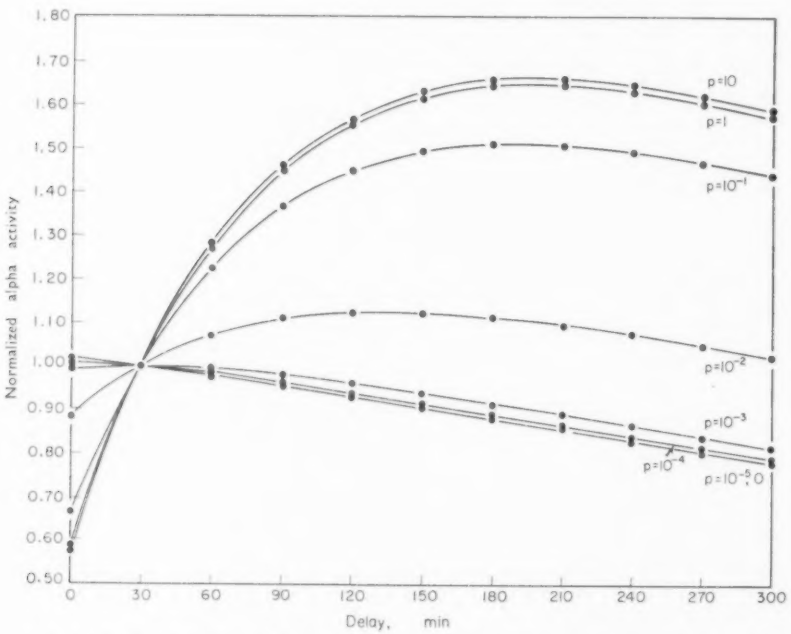


FIG. 1. α-Decay curves of filter-paper air samples for various degrees of ventilation.

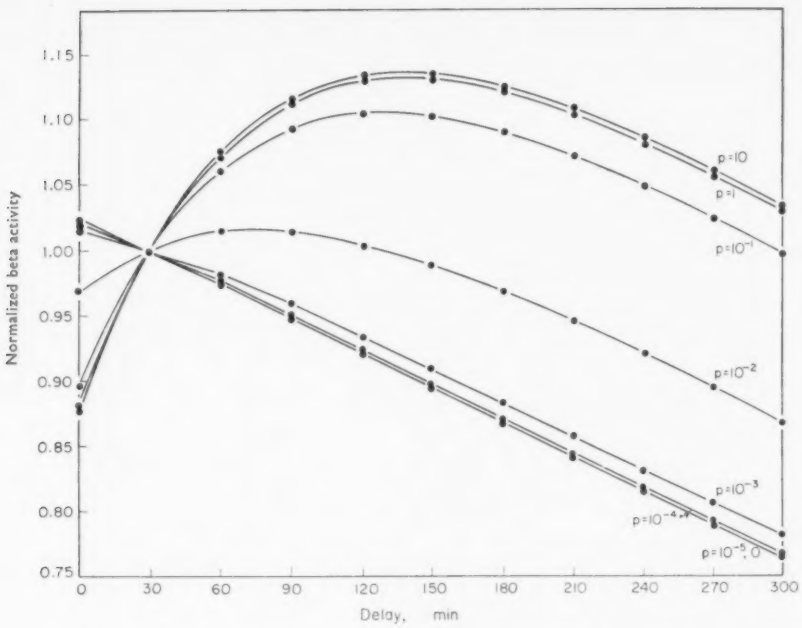


FIG. 2. β-Decay curves of filter-paper air samples for various degrees of ventilation.

across in practice. In addition, for any fixed change of ventilation rate, it will be observed that the shape of the curve changes more significantly in the case of α-decay than in the case of β-decay. In fact, this is what one would expect since the initial α-activity of the sample is primarily dependent on the number of ThC atoms present at the end of the sampling time and hence is susceptible to wide variations with respect to changes in p.

PRACTICAL APPLICATIONS OF THE METHOD

Since both the α- and β-activities of the samples from non-equilibrium atmospheres (ventilated places) nearly approach their maximum values at about $T = 150$ min, the ratio of the activities corresponding to $T = 150$ min and $T = 30$ min, (A_{150}/A_{30}), is chosen as a convenient parameter to be used in practice. Values for this ratio are calculated for different values of p and plotted in Fig. 3.

It is evident that the ventilation rate can be directly read out from these curves, once we know the value of A_{150}/A_{30}, which can be determined experimentally in any given case. It is easily seen that the measurement of α-activity gives a more accurate value of p than that obtained by a measurement of β-activity.

It is assumed in the above treatment that the radon concentration is negligible compared to that of thoron. It is also to be noted that ventilation affects the mean particle size of the dust in the atmosphere and this in turn affects the sample collection efficiency and losses due to self-absorption in the filter-paper. However, these factors do not lead to any error in the evaluation of p, the rate of ventilation. But the estimation of thoron concentration will be in error and the correct evaluation of this concentration is possible only if all these parameters are known for the particular rate of ventilation.

The following worked example illustrates the principles of the method: An air sample collected at the rate of 10 l/min for 1 hr using a filter-paper with 90 per cent efficiency of retention for the decay products of thoron, gives an α-activity of 1394 dis/min after a delay of 30 min (efficiency of detection of α-particles and self-absorption are taken into account). A value of 10^{-2} min^{-1} is obtained for p from Fig. 3.

The expected value of the activity for a thoron

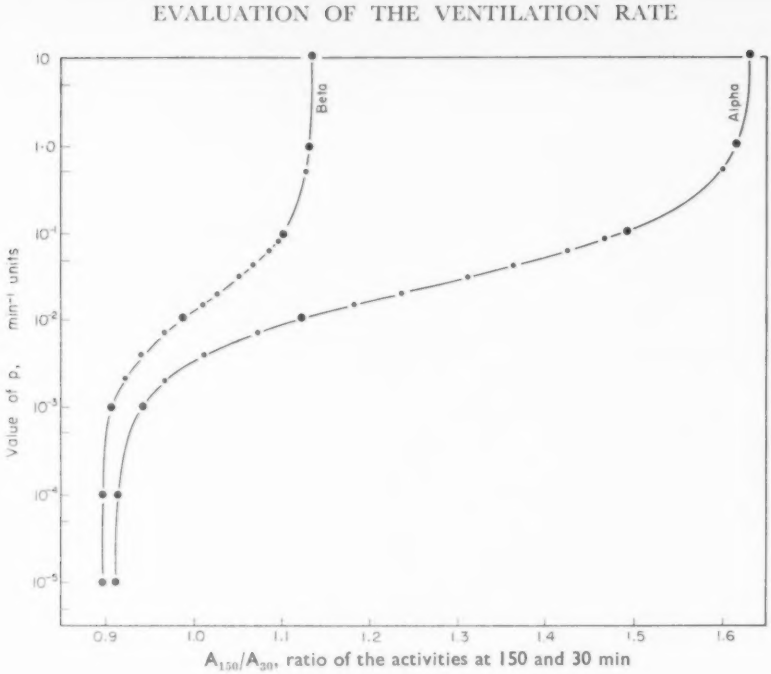


FIG. 3. Variation of A_{150}/A_{30} with p.

concentration of 1 atom per l. is 3.340 dis/min (Table 2). Therefore actual concentration of thoron

$$= \frac{1384}{0.9 \times 10 \times 3.340} \text{ atoms/l.}$$
$$= 1.58 \times 10^{-8} \mu c/cm^3 *$$

THE INHALATION HAZARD AS A FUNCTION OF THE VENTILATION RATE

The maximum permissible concentration (MPC) in air for thoron and radon is usually expressed in terms of $\mu c/cm^3$. Their daughter elements are assumed to be present in equilibrium concentrations; a condition which does not exist normally. As the biological damage is mainly due to the α-energy released, the maximum permissible level is sometimes considered in terms of the total α-energy resulting from the ultimate decay of all the daughter products associated with a certain volume of air.[12,13] A radon concentration of 10^{-7} $\mu c/cm^3$ in equilibrium with the daughter products is equivalent to 128 MeV total potential α-energy per cm^3 of air. The corresponding value for thoron and its daughters is 1750 MeV per cm^3. Thus, based on a consideration of potential α-energy alone, an equilibrium concentration of thoron is about 13.7 times more hazardous than an equivalent concentration of radon.

The potential α-energy values are calculated for a certain concentration of the parent gas for different degrees of ventilation and are then plotted as percentage of the equilibrium value against the ventilation rate in Fig. 4. It is seen from the figure that for the same percentage reduction of the potential α-energy, one needs higher rate of ventilation for radon than for thoron. This should be expected because the effective half-life of the α-emitters from thoron daughters is more than that of the α-emitters from radon daughters.

Acknowledgments—I am grateful to Mr. A. S. RAO for the encouragement and the guidance during this work. Thanks are due to Dr. E. C. TSIVOGLOU of the United States Public Health Service for useful comments.

* *Note added in proof:* The concentration works out to 1.67×10^{-8} $\mu c/cm^3$ if the more recent value of 51.5 sec is adopted for the half-life of thoron.

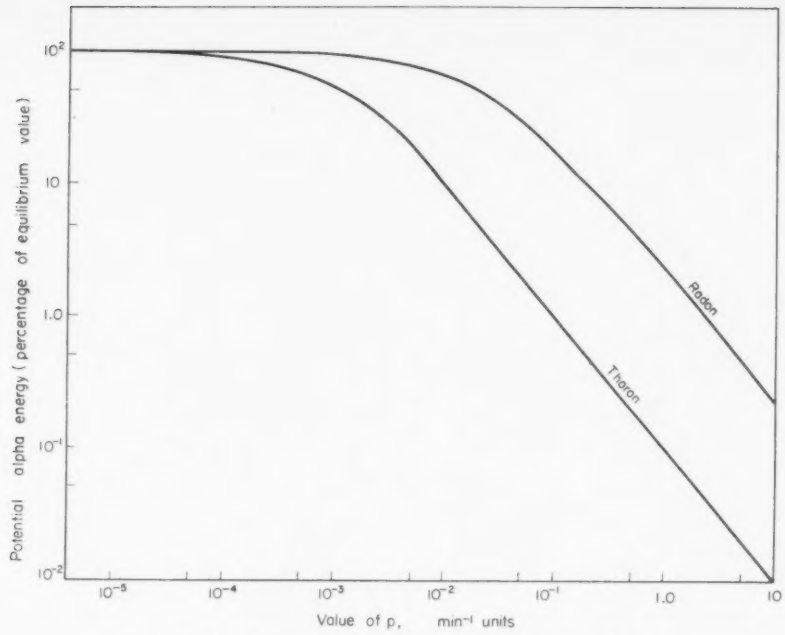


FIG. 4. Potential α-energy as a function of p.

REFERENCES

1. K. Z. MORGAN. Quoted E. C. TSIVOGLOU, in *A.M.A. Arch. Industr. Hlth.* **14,** 69. (1956).
2. A. C. CHAMBERLAIN and E. D. DYSON, *Brit. J. Radiol.* **29,** No. 342, 317 (1956).
3. S. H. COHN, R. K. SKOW and J. K. GONG, *Arch. Industr. Hyg.* **7,** 508 (1953).
4. J. SHAPIRO and W. F. BALE, University of Rochester Atomic Energy Project, UR-242.
5. J. SHAPIRO, *An Evaluation of the Pulmonary Radiation Dosage from Radon and Its Daughter Products.* University of Rochester Atomic Energy Project, UR-298 (1954).
6. W. G. BUSBRIDGE and A. C. CHAMBERLAIN, *Note on Radon Hazards in Uranium Mines.* AERE-HP/M-94.
7. D. A. HOLADAY, *A.M.A. Arch. Industr. Hlth.* **12,** 163 (1955).
8. E. C. TSIVOGLOU and H. E. AYER, *Arch. Industr. Hyg.* **10,** 363 (1954).
9. H. E. AYER, *Control of Radon and Its Daughters in Mines by Ventilation.* United States Atomic Energy Commission, AECU-2858 (1954).
10. E. C. TSIVOGLOU, H. E. AYER and D. A. HOLADAY, *Nucleonics* **11,** No. 9, 40 (1953).
11. B. HULTQVIST, *K. svenska Vetensk Akad. Handl.* **6,** 3 (1956).
12. H. L. KUSNETZ, *Amer. Ind. Hyg. Assn. Quart.* **17,** No. 1, 85 (1956).
13. D. A. HOLADAY *et al.*, *Control of Radon and Daughters in Uranium Mines and Calculations on Biological Effects.* Publication No. 494, U.S. Public Health Service (1957).

Health Physics Pergamon Press 1961. Vol. 6, pp. 142–148. Printed in Northern Ireland

MEASUREMENT OF CAESIUM-137 IN MILK

K. G. VOHRA, V. S. BHATNAGAR, U. C. MISHRA and S. RAGUPATHY
Air Monitoring Section, Atomic Energy Establishment, Trombay, Apollo Pier Road, Bombay 1, India

(*Received* 7 *July* 1960)

Abstract—Samples of liquid milk from Bombay and twenty-seven other stations in India, and imported powder milk from Australia, Denmark, Holland, U.K. and the United States have been measured for their caesium-137 and potassium-40 contents. The average caesium-137 content of Bombay milk is 9.38 $\mu\mu$c per g potassium for the year 1958, 18.26 $\mu\mu$c per g of potassium for 1959 and 14.71 $\mu\mu$c/g potassium for the first 2 months of 1960. The increase in the average level of caesium-137 by a factor of 2 during 1959 has been attributed to the accelerated fallout rate following the large number of tests conducted during the autumn of 1958. The caesium-137 content in the Australian powder milk is found to be about the same and in the European powder milk about two or three times that in the Indian powder milk. There are large variations in the levels of caesium-137 in different samples from India including Gangtok (Sikkim State). The values range from 2.79 to 159.9 $\mu\mu$c per g of potassium, the highest value being for Gangtok.

INTRODUCTION

Caesium-137 which is one of the long-lived radioactive products (half-life 30 years) in the fallout from the testing of nuclear weapons is disseminated on a world-wide scale by the stratospheric fallout and finds entry into biological materials, thus contributing to the biological hazard of fallout. The other long-lived radioactive products present in fallout in significant quantities are: strontium-90 (half-life 28 years), carbon-14 (half-life 5568 years) and plutonium-239 (half-life 24,360 years). The incorporation of any of these products in the biological cycle will depend on its chemical properties, and the associated biological hazard will depend on its biological retention in the human body and the type of radiation emitted. Caesium-137 is chemically similar to potassium and is therefore ingested quantitatively when taken into the animal body through food. The excretion of caesium-137 through milk is quite rapid, e.g. its biological half-life in cow is only 20 days. The estimation of caesium-137 in milk is useful because milk is the main diet of growing children. The ingested caesium-137 remains in the human muscle and its radiological effects are produced primarily by the 661 keV γ-emission causing whole body irradiation. The long-term hazard of caesium-137 is the gonadal dose received through whole body irradiation giving rise to genetic effects.

The natural potassium in milk is also radioactive because of the presence of natural isotope potassium-40 in it which emits 1.46 MeV γ-rays. The natural potassium-40 activity in milk ranges from 100 to 150 $\mu\mu$c per l. of liquid milk and from 0.5 to 1.5 $\mu\mu$c per g of powder milk. The variations in the natural radioactivity in milk can be used as a basis for assessing the biological hazard of caesium-137. This paper presents some data on the caesium-137 and potassium-40 levels in samples of milk from India and a few other countries. The data on natural potassium content of milk, and caesium-137 levels relative to potassium, are also presented. This data is considered useful in two ways, firstly in assessing the biological hazard of the present level of caesium-137 in milk, and secondly, in comparing the fallout in different countries.

SAMPLING

(a) *Bombay milk*

The sampling procedure is as follows: 2 l. of liquid milk are ashed daily and the ash collected over a period of one calender month is pooled

to get a sample large enough for accurate activity measurements. The ashing is carried out at a temperature of 400°C. The γ-activities due to caesium-137 (barium-137m) and potassium-40 are estimated using a scintillation spectrometer.

(b) *All India milk samples*

Twenty-seven sampling stations have been established in India (Fig. 1) under a programme of country-wide measurements recently undertaken. A sample of about 60 lb of liquid milk collected on a single day is received from each station every alternate month. The samples are received in polythene carboys and are ashed and measured.

(c) *Powder milk samples*

Powder milk samples from Australia, Denmark, Holland, India, United Kingdom and the U.S.A. were obtained from the market. Large quantities of powder milk are imported into the country and mainly used as baby food. The samples are either measured directly or in the form of ash, depending on the activity of the sample.

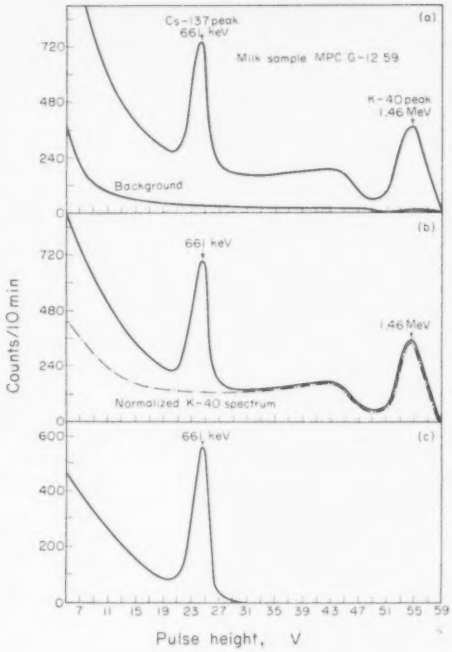


FIG. 1. Analysis of milk spectrum.

MEASUREMENT PROCEDURE

The measurements of potassium-40 and caesium-137 activity in milk are made using a 3 in. diameter × 1 in. sodium iodide scintillation detector with a ten-channel pulse-height analyser. The sample is placed in a specially designed container which is fixed on the sodium iodide crystal, giving a large counting geometry. The detector assembly along with the sample is placed in a $3\frac{1}{2}$-in. lead shield. The container can hold the ash from about 40 l. of milk.

The γ-ray spectrum of a typical milk sample obtained with the above set up is shown in Fig. 1(a). The background spectrum is also given in the same figure.

Fig. 1(b) gives the milk spectrum obtained after subtracting the background spectrum from the sample spectrum. The photopeaks corresponding to 0.661 MeV γ-rays from caesium-137, barium-137m and 1.46 MeV γ-rays from potassium-40 are marked in the spectrum. The following procedure is adopted for finding out the caesium-137 and potassium-40 content in milk ash. A potassium-40 spectrum is obtained using a standard potassium source. This spectrum is normalized with respect to the potassium-40 photopeak of the milk spectrum (as shown by the dotted line in Fig. 2(b). The normalized potassium-40 spectrum is subtracted from the milk spectrum. The resultant spectrum is due to caesium-137 (barium-137m) activity [Fig. 1(c)]. The actual magnitude of caesium-137 activity in the sample is found by comparing this with the spectrum of a standard caesium-137 source. Potassium-40 content of the samples is found by comparing the photopeaks of the sample and the standard source. The standard potassium-40 and caesium-137 sources have the same density as the sample and they are counted under the same geometrical conditions.

The bias of the spectrometer and the channel width are so adjusted that a complete sample spectrum is obtained in a single run of the pulse-height analyser. The background spectrum and the standard potassium-40 and caesium-137 spectra are obtained for each setting of the instrument for obtaining milk spectra.

RESULTS

Table 1 gives caesium-137 and potassium-40 γ-activities in liquid milk from Bombay for the different calender months of the period February 1958 to February 1960. Caesium-137

Table 1. *Caesium*-137 (*barium*-137*m*) *and potassium*-40 γ-*activity levels, and potassium content of Bombay milk*

Sample no.	Period of collection	Cs^{137}/g of potassium ($\mu\mu$c)	Cs^{137}/l. of milk ($\mu\mu$c)	K^{40}/l. of milk ($\mu\mu$c)	Potassium/l. of milk (g)
	1958				
M-6/58	February	9.10 ± 1.34	17	181	1.83
M-8/58	March	10.72 ± 2.63	10	94	0.95
M-9/58	April	7.00 ± 1.20	11	158	1.60
M-10/58	May	9.16 ± 1.45	9	95	0.96
M-11/58	June	4.83 ± 0.96	5	98	0.99
M-12/58	July	17.20 ± 2.74	10	58	0.59
M-13/58	August	9.31 ± 1.40	13	138	1.40
M-14/58	September	11.65 ± 1.00	17	143	1.45
M-16/58	November	9.90 ± 1.60	10	99	1.00
M-17/58	December	4.93 ± 1.00	5	102	1.03
	1959				
M-18/59	January	10.15 ± 1.56	12	118	1.20
M-19/59	February	22.90 ± 0.86	21	91	0.92
M-20/59	March	9.33 ± 1.47	11	114	1.15
M-21/59	April	14.40 ± 0.75	13	86	0.87
M-22/59	May	14.50 ± 0.74	12	80	0.81
M-23/59	June	9.98 ± 0.70	8	76	0.77
M-24/59	July	23.95 ± 0.62	18	73	0.74
M-25/59	August	33.15 ± 0.89	27	80	0.81
M-26/59	September	28.10 ± 0.93	19	68	0.69
M-27/59	October	20.65 ± 0.85	19	92	0.93
M-28/59	November	16.48 ± 0.65	16	97	0.98
M-29/59	December	15.60 ± 0.77	12	77	0.78
	1960				
M-30/60	January	14.17 ± 0.73	12	84	0.85
M-31/60	5 February	17.15 ± 0.79	23	132	1.34
M-32/60	20 February	12.80 ± 0.88	14	106	1.07
Average	Year 1958	9.38 ± 1.65	11	117	1.18
Average	Year 1959	18.26 ± 0.95	16	88	0.89
Average	Year 1960	14.71 ± 0.80	16	107	1.09

activity of the samples is expressed as micro-microcuries ($\mu\mu$c) γ-activity per gramme of potassium. Table 2 gives the data for liquid milk samples from the different sampling stations in India. Table 3 gives the data for the powder milk samples from Australia, Denmark, Holland, India, United Kingdom and the U.S.A.

DISCUSSION

The average caesium-137 content of the Bombay milk is about 9.4 units ($\mu\mu$c/g potassium) for 1958, and about 18.3 units for the year 1959 which is nearly twice the 1958 level. Anderson *et al.*[1] have suggested that caesium-137 in milk may be a direct measure of the fallout rate due to its short biological half-life in cow.[2] Accordingly, one may conclude that the fallout rate during 1959 has been twice the average fallout rate for 1958. The increase observed in 1959 may be attributed to the increased fallout rate following the large number of tests conducted during the autumn of 1958.

Table 2. Caesium-137 (barium-137m) and potassium-40 γ-activity levels, and potassium content of milk samples from different stations in India

Place	Location	Sample no.	Date of collection	Cs^{137}/g of potassium ($\mu\mu$c)	Cs^{137}/l. of milk ($\mu\mu$c)	K^{40}/l. of milk ($\mu\mu$c)	Potassium/l. of milk (g)
Adoni	15.5·N, 77°E	AN-1	19-9-59	12.98 ± 0.47	11	80	0.81
Adoni	15.5·N, 77°E	AN-2	3-2-60	8.61 ± 0.57	6	71	0.72
Allahabad	25.22N 81.50E	AL-1	13-11-59	13.46 ± 0.51	16	116	1.18
Anand	22.5N 73E	AD-1	20-2-60	5.10 ± 0.41	5	92	0.93
Bangalore	12.45N 77.3E	BA-1	12-8-59	10.85 ± 0.44	14	130	1.32
Bangalore	12.45N 77.3E	BA-2	16-2-60	2.54 ± 0.32	29	112	1.13
Bhavnagar	22N 72E	BH-1	21-8-59	9.07 ± 0.32	11	119	1.21
Bhavnagar	22N 72E	BH-2	16-11-59	5.80 ± 0.35	7	123	1.25
Bikaner	28N 73.3E	BK-1	30-1-60	4.31 ± 0.39	5	119	1.21
Calcutta	22.3N 88.3E	CA-1	20-2-60	2.20 ± 0.28	3	115	1.17
Coimbatore	11N 77E	CB-1	14-11-59	4.13 ± 0.26	6	151	1.53
Cuttack	22.3N 86E	CK-1	20-8-59	8.90 ± 0.33	9	96	0.97
Cuttack	22.3N 86E	CK-2	20-11-59	7.65 ± 0.52	7	84	0.85
Guntur	16.5N 80.5E	GU-1	22-8-59	7.65 ± 0.32	7	98	0.99
Jabalpur	23.15N 80E	JA-1	23-1-60	3.06 ± 0.46	3	94	0.95
Kanpur	26.3N 80.15E	KP-1	20-8-59	9.49 ± 0.34	10	100	1.01
Kanpur	26.3N 80.15E	KP-2	17-11-59	3.20 ± 0.42	3	90	0.91
Karnal	29.7N 77E	KL-1	19-8-59	14.70 ± 0.41	18	121	1.23
Kodaikanal	10N 78E	KK-1	6-11-59	4.04 ± 0.52	3	73	0.74
Kodaikanal	10N 78E	KK-2	18-1-59	13.20 ± 0.47	16	119	1.21
Lucknow	26.45N 80.5E	LW-1	3-11-59	11.31 ± 0.58	10	87	0.88
Madras	13N 80.3E	MA-1	14-12-59	13.10 ± 0.51	14	103	1.04
Meerut	28.45N 78E	MT-1	4-12-59	5.83 ± 0.45	5	89	0.90
Muzaffarpur	26.2N 85.5E	MP-1	19-2-60	2.79 ± 0.30	3	115	1.17
Mercara	12.5NZ 75.6E	MC-1	28-12-59	3.61 ± 0.33	4	115	1.17
Patna	25.3N 85.15E	PA-1	26-8-59	5.32 ± 0.26	5	88	0.89
Patna	25.3N 85.15E	PA-2	10-12-59	5.20 ± 0.49	4	79	0.80
Poona	18.3N 74.0E	PO-1	20-8-59	21.82 ± 0.87	21	93	0.94
Poona	18.3N 74.0E	PO-2	12-1-60	4.60 ± 0.47	4	84	0.85
Sikkim	27.2N 88.4E	SK-1	13-12-59	56.85 ± 2.24	138	240	2.43
Sikkim	27.2N 88.4E	SK-2	13-12-59	159.90 ± 6.39	262	162	1.64
Sikkim	27.2N 88.4E	SK-3	13-12-59	21.33 ± 1.43	22	100	1.01
Shillong	25.6N 91.9E	SH-1	10-9-59	12.64 ± 0.56	24	187	1.89
Shillong	25.6N 91.9E	SH-2	12-10-59	17.50 ± 0.75	27	155	1.57
Trivandrum	8.5N 77E	TV-1	27-11-59	14.32 ± 0.48	18	123	1.25
Varanasi	24.3N 83.2E	VR-1	30-11-59	3.49 ± 0.42	3	86	0.87
Visakhapatnam	17.5N 83.3E	VK-1	1-11-59	12.48 ± 0.69	9	75	0.76

Fig. 2 gives the monthly levels of caesium-137 ($\mu\mu$c/g of potassium) in Bombay milk for the period February 1958 to February 1960. This figure shows significant peaks in caesium-137 activity during three different periods, i.e. July 1958, February 1959 and August 1959. The increase in the months of July 1958 and August 1959 may be explained to be due to the accelerated deposition by heavy rainfall during the monsoon months.[3] The summer monsoons at Bombay during which there is heavy precipitation prevail from June to September. The increase in activity observed during the period January to March 1959 coincides with the

Table 3. Caesium-137 (barium-137m) and potassium-40 γ-activity levels, and potassium content of powder milk

Country	Sample no.	Date of receipt	Cs^{137}/g of potassium ($\mu\mu c$)	Cs^{137}/kg of milk powder ($\mu\mu c$)	K^{40}/kg of milk powder ($\mu\mu c$)	Potassium/kg of milk powder (g)
Australia	MPN-10/59	Mar. 1959	4.02 ± 0.85	51	1256	12.73
Australia	MPL-18/59	Sept. 1959	4.56 ± 1.33	27	587	5.95
Australia	MPN-25/59	Oct. 1959	10.79 ± 1.47	138	1261	12.78
Denmark	MPM-22/59	Oct. 1959	17.60 ± 2.09	263	1471	14.90
Europe	MPD-6/58	Dec. 1958	19.75 ± 1.68	330	1648	16.70
Europe	MPD-19/59	Sept. 1959	34.00 ± 3.30	447	1298	13.15
Holland	MPO-2/58	July 1958	20.00 ± 3.00	288	1421	14.40
Holland	MPK-5/58	Dec. 1958	22.15 ± 1.96	300	1337	13.55
Holland	MPO-12/59	Apr. 1959	23.55 ± 2.85	281	1177	11.92
Holland	MPO-16/59	Sept. 1959	18.92 ± 2.22	325	1698	17.20
Holland	MPK-17/59	Sept. 1959	13.82 ± 1.96	214	1530	15.50
India	MPA-3/58	Dec. 1958	5.10 ± 1.17	48	935	9.47
India	MPA-7/59	Jan. 1959	8.33 ± 1.45	83	982	9.95
India	MPA-8/59	Feb. 1959	7.07 ± 1.59	71	997	10.10
India	MPA-13/59	Mar. 1959	17.40 ± 3.82	154	872	8.83
U.K.	MPCG-9/59	Mar. 1959	31.60 ± 2.62	347	1084	10.98
U.K.	MPOS-14/59	June 1959	24.10 ± 2.50	295	1209	12.25
U.K.	MPG-15/59	June 1959	23.50 ± 4.10	174	730	7.40
U.S.A.	MPS-21/59	Oct. 1959	19.30 ± 2.15	262	1337	13.55

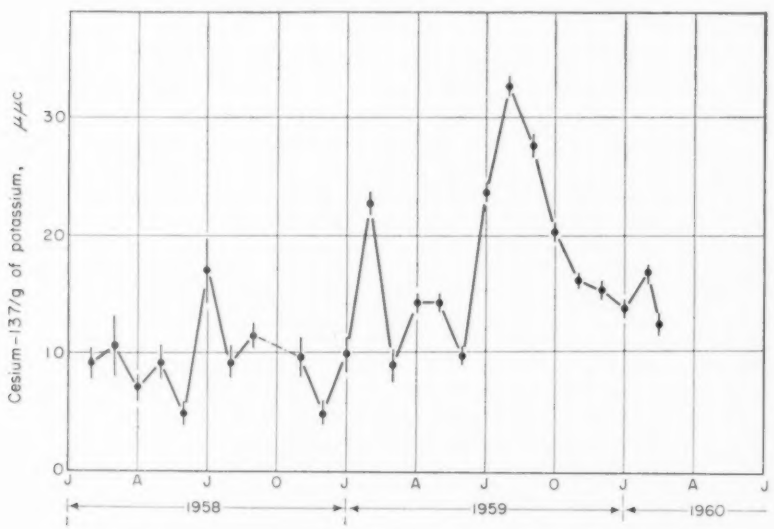


FIG. 2. Average monthly levels of caesium-137 in Bombay milk ($\mu\mu c$/g potassium) during the period 1958–1960.

increase in airborne activity during the same period.[4] The general nature of variations of caesium-137 in milk is similar to the variations observed in the strontium-90 level in milk at Bombay.[5]

The average values (in micromicrocuries per gramme of potassium) of caesium-137 content in the samples from the different stations in India and from Gangtok (Sikkim State) are charted on the map in Fig. 3.

There are large variations in the values at different stations, and the value obtained at Gangtok (6000 ft altitude station) is by far the highest i.e. 262 $\mu\mu$c per l. of milk (Table 2), nearly ten times the average value for all the stations in India. The variations may be expected to be due to the differences in rainfall giving different degrees of fallout deposition. The other factors determining the degree of deposition are the latitude and altitude of the station and the nature of prevailing winds. The variations may also be due to the type and the source of the pasture on which the animal is fed. The high value at Gangtok is likely to be due to a combination of these several factors.

A comparison between the caesium-137 content in the Indian, European and the Australian powder milk shows that the content in the

FIG. 3. Average levels of caesium-137 in milk ($\mu\mu$c/g potassium) from different places in India, during 1959–1960.

Australian milk is about the same and in the European milk about two to three times that in the Indian milk. The variations give an indirect indication of the general fallout rates in the regions. This cannot be a precise statement because the samples may not correspond to the same periods in all the cases and the composition of the different powders may not be the same.

The activity levels of caesium-137 in most of the samples studied are only a small fraction of the natural potassium-40 γ-activity in these samples. For example the caesium-137 (barium-137 m) γ-activity in Bombay milk is only about 10 per cent of the corresponding potassium-40 γ-activity. The normal variations in the potassium-40 activity of milk are often much larger than this, and therefore, the present levels of caesium-137 in milk may not be of much significance as a health hazard. Even the highest value at Gangtok is only a small fraction of the tolerance level.

Acknowledgments—Our thanks are due to SHRI A. S. RAO for his guidance and constant interest in the present work. Our thanks are also due to the Ministry of Food and Agriculture, Government of India, for arranging the supply of milk samples.

REFERENCES

1. E. C. ANDERSON, *Science* **125,** 1273 (1957).
2. S. L. HOOK and C. L. COMAR, University of Tennessee Report ORO-91 (1953).
3. K. G. VOHRA, C. RANGARAJAN and M. C. JAIN, *Indian J. Met. Geophys.* **11,** 117 (1960).
4. K. G. VOHRA, C. RANGARAJAN and N. A. INGLE, *Indian J. Met. Geophys.* **11,** 123 (1960).
5. K. G. VOHRA, C. W. JOSHI, S. B. HINGORANI and B. Y. LALIT, *Indian J. Med. Sci.* **14,** 811 (1960).

Health Physics Pergamon Press 1961. Vol. 6, pp. 149–151. Printed in Northern Ireland

A PROPORTIONAL CONTINUOUS RIVER AND STREAM WATER SAMPLER AT GEORGIA NUCLEAR LABORATORIES

J. M. SELBY*, J. R. WRIGHT, B. M. BOWEN and J. H. EDGERTON
Nuclear Safety Department, Lockheed Nuclear Products, Lockheed Aircraft Corporation, Georgia Division, Marietta, Georgia

(*Received* 15 *November* 1960)

Abstract—The main facility at the Georgia Nuclear Laboratories (GNL) is an air-shielded 10 MW reactor designed for a high leakage flux. This reactor, located within 750 ft of the Etowah River, is used for irradiation of aircraft systems and components. Because of the high neutron dose and the resultant soil activation, rigid control is maintained on run-off streams and the Etowah River. In order to overcome the lack of power at river and stream sampling sites, a sampler was designed to utilize river power. The sampler, built on the water wheel principle, includes an adjustable suction device and a revolution counter. Using a current measuring apparatus, such as a Gurley meter, and maintaining the sampler at a desired sampling rate, the total revolutions and the volume collected can be related to the rate of flow of the river. The construction of the continuous proportional water sampler and the calibration are discussed in this report.

INTRODUCTION

The Georgia Nuclear Laboratories (GNL), located 50 miles north of Atlanta,[1] is operated by Lockheed Nuclear Products for the United States Air Force. The original concept of the laboratory was to provide a facility where large aircraft systems and components could be dynamically tested and evaluated under realistic flux and environmental conditions. The facility is also used in ecological, biomedical and shielding studies.

In order to irradiate large systems, it was necessary to have a wide radiation field with a high flux throughout. The Radiation Effects Reactor (RER) was designed by the Atomic Power Equipment Department of General Electric Company, San Jose, California, from a conceptual design by Lockheed's Nuclear Branch to accomplish that purpose. The RER is a heterogeneous, pressurized, light water moderated and cooled reactor having a core of twenty-six modified MTR type fuel elements, four fuel–cadmium poison control rods, and a cadmium–silver regulating rod. The reactor, designed to operate at 10 MW with a fast neutron leakage of 2.8×10^{11} n/cm^2 sec[2] is supported on a hydraulic lift. The RER is raised to a position above a 38-ft deep storage pool and operated remotely from a control room in an adjoining underground concrete building.

SAMPLER REQUIREMENT

Since the Radiation Effects Reactor is operated above ground and is air shielded, a high neutron flux exists in the area surrounding the reactor. The flux in n/cm^2 per sec at varying distances from the RER, as measured by the activation of manganese specimens,[3] is shown in Fig. 1. Activation occurs in the soil as a result of the escape neutrons from the RER. The gross β–γ activation versus distance relationship is shown in Fig. 2. The reactor is located 750 ft from the Etowah River, which is a source of water for several downstream communities. The gross radioactivity density in run-off water resulting from erosion of activated soil has been measured as high as 1.2×10^{-6} μc/cm^3. In order to insure that the operation of the RER and subsequent erosion of activated soil creates no problem to GNL or off-site personnel, it was necessary to maintain close observation of radioactivity concentrations in the effluent river

* Present Address: General Electric Co., Hanford Atomic Products Operation, Richland, Washington

water. Eighteen grab water samples are collected weekly from the Etowah and two tributaries within the plant boundary.[4] These sample sites (Fig. 3) were established for background purposes above the reactor and at locations where activity might concentrate below the reactor. However, to provide a continuous record of the radioactivity density in the river it was necessary to collect a continuous proportional sample.

SAMPLER CONSTRUCTION

Several continuous water sample systems are listed in the literature. An electrically operated pump was installed on the Columbia River by the General Electric Company.[5] Oak Ridge National Laboratory has an electrical system that continuously samples the effluent at the White Oak Creek Dam.[6] Metered siphon samplers have been reported by several sources and are currently used by the Georgia Public Health Department in sampling the Etowah River below the plant.

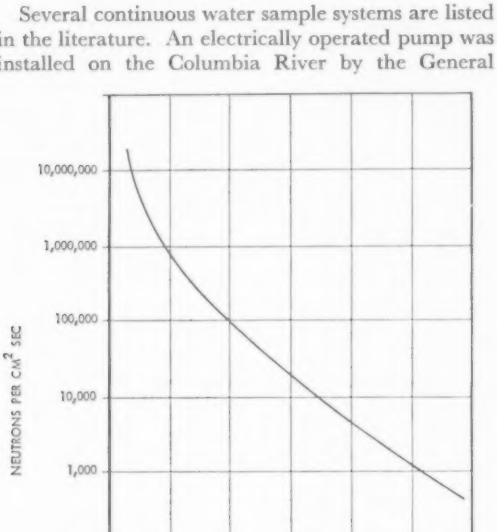


FIG. 1. Neutron flux vs. distance at 10 MW.

It was the opinion of GNL personnel that a proportional sample should be collected to give integrated results, thereby eliminating siphon-principle samplers.

The locations chosen for continuous samplers (Fig. 3) were in remote areas where no electric power

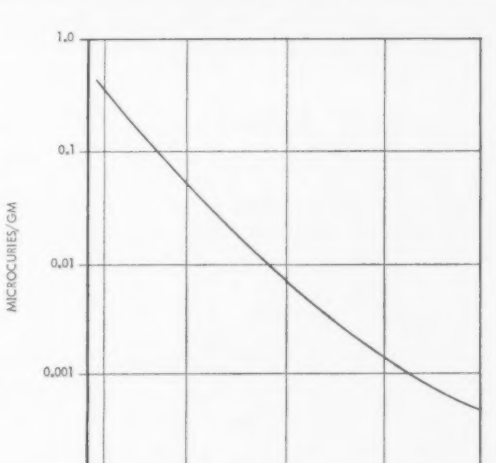


FIG. 2. Activation vs. distance from RER after 21 hr at 10 MW.

was available. Therefore a sampler had to be designed that would use the river as a source of power.

The design consists of a 24-in. aluminum water wheel suspended from a floating platform secured to river bank by cables. The wheel turns a cam which operates a standard laboratory double acting rubber bulb. With this arrangement a sample can be taken from a depth of 3 ft below and pumped 3 ft above the surface of the water being sampled. The cam arrangement can be adjusted from $\frac{1}{10}$ ml per wheel revolution to 15 ml per revolution. The design

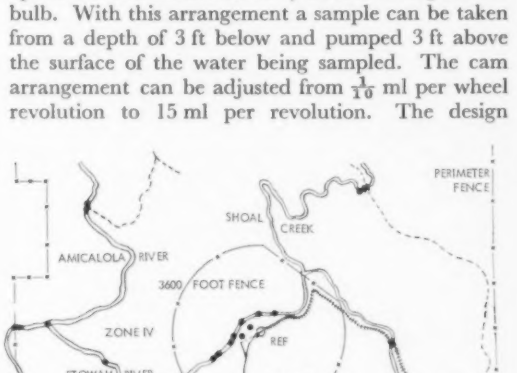


FIG. 3. River sampling sites.

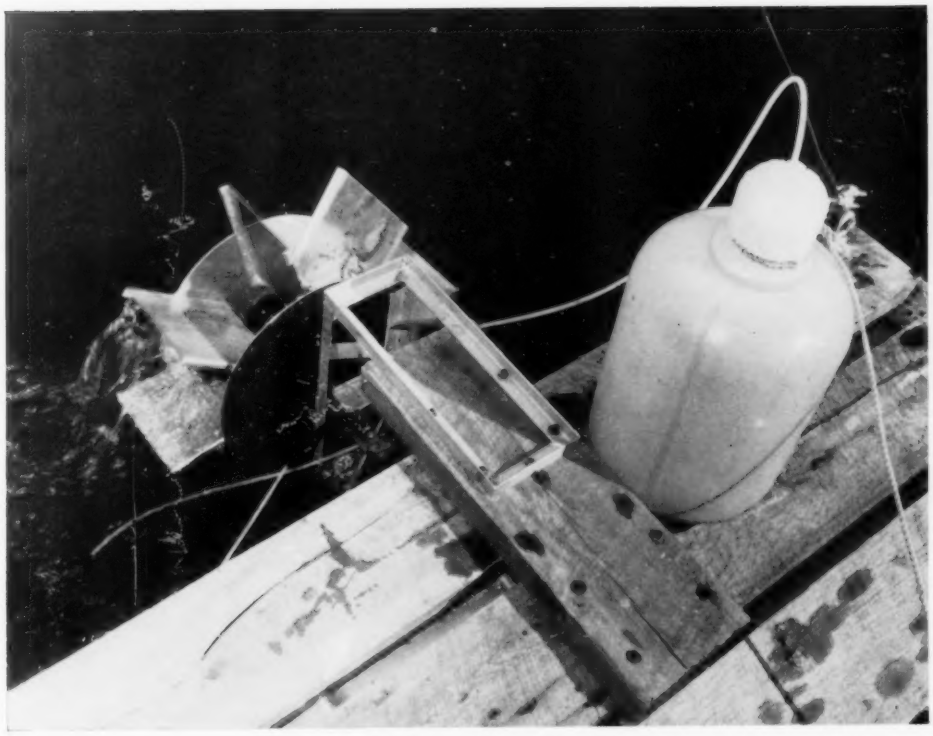


FIG. 4. Continuous river water sampler.

BLANK
PAGE

includes a revolution counter used in the calculation of the total volume collected and in the determination of the fluctuation in river flow rate. One of the continuous water samplers is shown in operation in Fig. 4.

In addition to the present measurements made with these samplers, it is planned to include ion-exchange columns so that much of the laboratory processing time may be eliminated.

OPERATING EXPERIENCE

Six continuous water samplers have been installed on-site to date. Several of these samplers have been operated for a period of 4 months with a minimum of maintenance required. During this period temperatures have been as low as 6°F without the samplers freezing.

The gross β–γ concentrations in grab water samples have been compared to those in the continuous samples during the first 9 weeks in 1960. Fig. 5 shows that the concentration in the continuous samples were consistently higher than the concentration in grab samples with the exceptions of the fourth and ninth weeks. During the fourth and ninth weeks excessive rains were experienced with an increase in run-off of irradiated soil. The grab samples were collected during the period of maximum run-off, thus more activity was present in these than in the integrated samples. Excluding these 2 weeks the average radioactivity concentration ratio of continuous to grab samples was 2.1 ± 0.87.

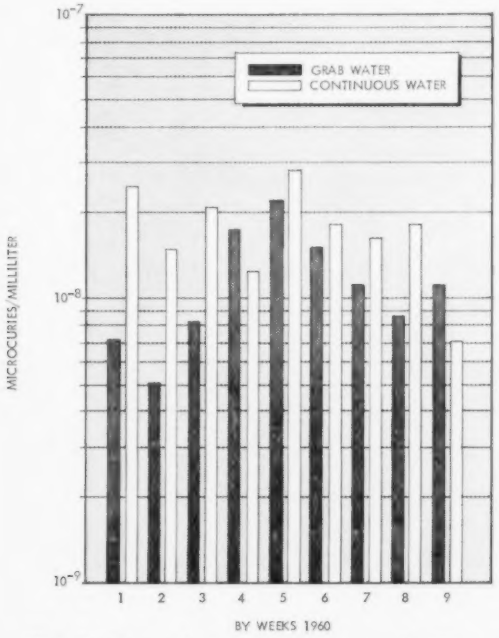


FIG. 5. Comparison grab water to continuous water sampler.

At the present time the sampler is proportional only to the linear flow of the river. However, with a river profile study and flow study at each sampler location and a river stage recorder, it will be possible also to determine total volume flow during each sampling period.

Acknowledgment—The assistance of J. E. BRAMBLETT, G. G. DOWNING, S. D. JOHNSON, W. T. PRICE, K. W. SMITH and N. W. WHITE in the preparation of this paper is gratefully acknowledged.

REFERENCES

1. *Radiation Effects Reactor Safeguards*, LAC 147, 15 January (1958).
2. J. H. EDGERTON, *Analytical Requirements of the Lockheed Critical Effects Reactor, of the Radiation Effects Reactor, and of the Radioactive Waste Disposal System*. AEC TID 7568, Pt. 1, pp. 50–74, February (1959)
3. Unpublished data, Nuclear Safety Department, Lockheed Nuclear Products Branch, Dawsonville, Georgia.
4. J. M. SELBY, B. M. BOWEN and J. H. EDGERTON, *Environmental Monitoring at Georgia Nuclear Laboratories-AFP* 67. Presented at the 21st Annual Meeting of the American Industrial Hygiene Association, Rochester, New York, 25–28 April (1960).
5. W. B. SILKER, *A Continuous River Sampling System and Methods of Radiochemical Analyses for Strontium-89, 90, Barium-140, and Phosphorus-32*. HW47578, 14 January, (1957).
6. J. H. EDGERTON. Private communication from J. C. HART.

Health Physics Pergamon Press 1961. Vol. 6, pp. 152–162. Printed in Northern Ireland

STATISTICAL TREATMENT OF ENVIRONMENTAL RADIO-ASSAY DATA

GERALD W. KERR
Industrial Hygiene and Safety Engineering Unit, Health and Safety Section,
Pratt & Whitney Aircraft, Division of United Aircraft Corporation,
Connecticut Operations-Canel, Middletown, Connecticut

(*Received* 11 *December* 1958)

Abstract—A program for environmental monitoring is outlined in this paper. The program described herein is applied to the CANEL site located at Middletown, Connecticut and operated for the U.S. Atomic Energy Commission by Pratt & Whitney Aircraft Division of United Aircraft Corporation. Information on sample locations, sampling methods and procedures, treatment and analysis of data and interpretation of data is given. Samples of water, soil, silt, vegetation, drinking water, air, precipitation and fallout are collected and analyzed. This program is offered as a new approach to environmental monitoring with respect to selection of sample locations, determination of sampling frequency and analysis of data.

A. INTRODUCTION

This report outlines the CANEL site monitoring program as it is being conducted at present. It provides information on background radiation present in the environment, both on and off site, and methods for noting changes in environmental activity. The program may be expanded in the future or modified to meet changing conditions. Larger facilities, more equipment and additional personnel will be required to carry out the program to its fullest extent, but a large portion of it is under way at the present time.

B. OBJECTIVES

The three basic objectives for the CANEL site environmental survey program are as follows:

(1) To accumulate data which may be analyzed by statistical methods and provide a sensitive comparison of pre-operational data and post-operational data at CANEL.

(2) To provide a mechanism for maintaining a running comparison of current data with all previous data or any portion of previous data to detect quickly trends or significant changes which might be due to operations conducted at CANEL. Statistical procedures alone are not the basis for interpretation of data. Interpretation is made upon the basis of statistical analyses, meteorological data, facility operations, and stack monitor data. Statistical analyses, however, give the first indication of significant increases in activity.

(3) To determine annual average concentrations of air and water activities for comparison with maximum permissible concentrations.

C. SAMPLING LOCATIONS

The general site and surrounding area is divided into circular sectors, as shown in Fig. 1. The circles are not equally spaced and, in fact, are at $\frac{1}{4}$, $\frac{1}{2}$, 1, 2, 3, 4, and 5 mile radii, respectively. Each annulus is denoted by the first number; i.e. from 1 to 7, inner ring to outer ring respectively.

The sectors are denoted by a second number as follows: northerly, 1; westerly, 2; southerly, 3; and easterly, 4. The directions and boundaries of the sectors were chosen by studying the wind roses for the area and in consultation with Weather Bureau personnel.

From 15 March 1956 to 31 May 1958 the U.S. Weather Bureau conducted a micrometeorological study at the CANEL site. This study is now being continued by Pratt & Whitney Aircraft personnel and covers the general climatology and diffusion climatology of the site. From a study of this data, it was

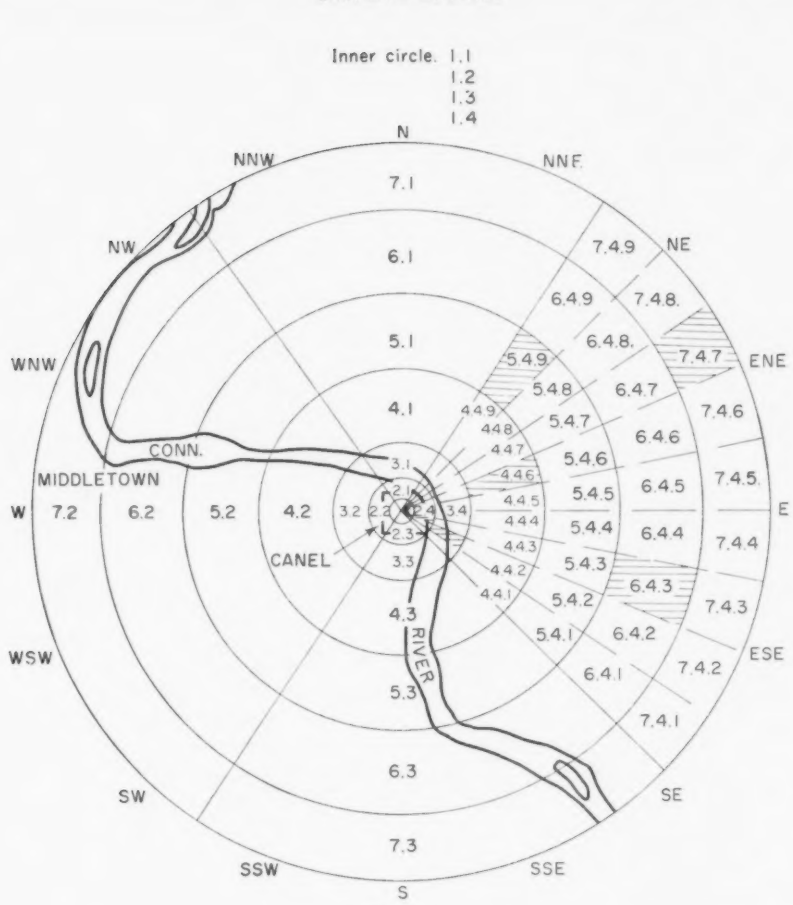


FIG. 1.

concluded that maximum ground level concentrations are expected to occur within the area contained by the 5-mile radius. It is believed that samples gathered at great distances from CANEL would be of little value, as there are at least four other nuclear installations within 50 miles of this site.

It is not necessary for the annuli and/or the sectors to be symmetrical. When data are analyzed and if, for example, it is found that distance is an influencing factor, basic data can be examined and the annulus found that is showing a significant increase in activity. It is imperative, however, that a sample be taken in each sector of each annulus in every sampling period.

It would be pertinent to mention at this point some findings of Weather Bureau personnel. In their preliminary smoke studies, it was noted that smoke was readily channeled up two ravines north of the site. The ravines follow the course of Mine Brook north of Middle Haddam and Great Hill Pond Brook which passes close to Cobalt. This channeling effect is noticeable when a south wind is prevalent. The effect is also evident, but to a lesser extent, southward

along the Connecticut Valley when a north wind prevails. A permanent monitoring station will be placed in one of the north ravines and in the valley to the south of the site. Further study will be made regarding the establishment of these permanent stations. Although not an integral part of the environmental survey program, liquid wastes and stack effluents will be monitored to prevent release of excessive concentrations of activity to the environment.

D. SAMPLING FREQUENCY AND INCIDENTS

As this section deals largely with statistical processes, a statement of terminology is necessary. For this section alpha (α) and beta (β), Greek letters, will be used to indicate types of statistical errors and should not be confused with α- and β-radiation.

Terminology

$H:$ = a stated hypothesis;

$\bar{x} = \dfrac{x_1 + x_2 \ldots\ldots + x_n}{n}$ = mean of a sample of size n;

$\bar{\bar{x}} = \dfrac{\bar{x}_a + \bar{x}_b + \ldots\ldots \bar{x}_k}{k}$ = mean of a group of sample means;

$s_n^2 = \sum_{i=1}^{n} \dfrac{(x_i - \bar{x})^2}{n-1}$ = best estimate of variance of a sample of size n;

$s_p^2 = \dfrac{(n_1-1)s_1^2 + (n_2-1)s_2^2 + \ldots\ldots + (n_k-1)s_k^2}{N-k}$ = pooled estimate of variance for a group of samples each of size $n_1, n_2, \ldots\ldots, n_k$;

n = each sample size;

k = number of groups of samples;

N = total number of samples;

m = degrees of freedom of a preliminary sample set $= N_1 - 1$;

$d.f.$ = degrees of freedom of a second sample set $= N_2 - 1$;

μ_0 = assumed number and in this case numerically equal to $\bar{\bar{x}}$;

μ = true mean of a universe (a set of objects having a common observable characteristic);

a = quantity numerically equal to $\mu - \mu_0$;

α = level of significance and is the chance of deciding that the hypothesis (such as $\mu = \mu_0$) is false when in fact the hypothesis is true;

β = chance of deciding a hypothesis (such as $\mu = \mu_0$) is true when in fact the hypothesis is false;

$1\text{-}\beta$ = power of a test.

The α- and β-errors can be illustrated by the following scheme:

		The decision	
		$H:$ true	$H:$ false
True state of nature	$H:$ true	Correct	α-error
	$H:$ false	β-error	correct

To determine the number of samples to be taken in a given period of time and obtain statistically valid results, it is necessary to use some data from past experience.

Taking data from 1956 off-site silt samples which had homogeneity of variance

$$s_p = 68.7$$
$$\bar{\bar{x}} = 60 \ \mu\mu c/g$$
$$N_1 = 30$$

Assuming it is desirable to know when the value 100 $\mu\mu$c/g is reached, the problem is to find N_2 to test

H: $\mu = \mu_0 = 60$

with: $\alpha = 0.05$

$\beta = 0.20$

$1 - \beta = 0.80$

This means there is a chance 0.80 of rejecting H: $\mu = \mu_0 = 60$ if actually $\mu = 100$. Then:

$$\mu = \mu_0 + a = 60 + (100 - 60) = 60 + 40$$

and $a = 40$

$$m = 30 - 1 = 29$$

Calculate: $\dfrac{a}{s_p} = \dfrac{40}{68.7} = 0.583$

Entering the table shown in Fig. 2 for determination of a second sample size, with the values of $m = 29$, find 0.583 in column marked $m = 29$ (or interpolate) and it is found that $d.f. = 20$; thus $N_2 = 21$.

If data from preliminary water samples has a variance compatible with that shown above and a mean less than 60 $\mu\mu$c/l. (which is true for 1956 data) and it is desired to detect 100 $\mu\mu$c/l., which is the MPC of radioactive contaminants for use beyond the control area as stated in NBS Handbook 52,[1] the $N = 21$ would be on the safe side as far as reliability in results. Any increase in a above results in a decrease in N. It is desirable to make $N = 24$ for water, soil, vegetation and well water samples and $N = 16$ for silt samples. The first-mentioned samples are collected twice monthly for 12 months and silt samples twice monthly for 8 months. Thus, these values of N provide for more efficient use of the time of the sampling technician. This method of determining sample size does not stipulate over what period of time the samples may be taken. The number of samples, N_2, may be distributed over *a period of time*. The justification of 1 year as the period of time stems from the fact that the AEC allows air and water concentrations to be averaged over a period not greater than 1 year. This thinking was extrapolated from air and water to soil, vegetation and silt samples. The $N = 24$ does not hold good for vegetation samples if it is desired to detect a level of 100 $\mu\mu$c/g, as the activity in vegetation is considerably higher than that. There are no published values for an MPC in vegetation, as there can be no control of the

$d.f.$ \ m	1	2	3	4	5	6	8	12	16	24	32	∞
1	13.8	8.53	7.39	6.93	6.68	6.51	6.31	6.13	6.04	5.96	5.92	5.79
2	5.88	3.51	3.02	2.81	2.70	2.62	2.53	2.45	2.41	2.37	2.35	2.30
3	4.30	2.55	2.20	2.03	1.96	1.91	1.85	1.78	1.75	1.72	1.70	1.65
4	3.55	2.10	1.80	1.67	1.60	1.56	1.50	1.45	1.43	1.40	1.39	1.36
5	3.12	1.85	1.58	1.47	1.41	1.37	1.32	1.28	1.25	1.23	1.22	1.18
6	2.81	1.66	1.43	1.32	1.27	1.23	1.19	1.15	1.13	1.11	1.10	1.07
7	2.56	1.52	1.30	1.21	1.16	1.12	1.08	1.05	1.03	1.02	1.01	0.979
8	2.37	1.41	1.21	1.12	1.07	1.04	1.00	0.972	0.956	0.940	0.932	0.910
9	2.23	1.32	1.14	1.05	1.01	0.978	0.944	0.913	0.898	0.883	0.875	0.854
10	2.11	1.25	1.07	0.993	0.952	0.925	0.893	0.863	0.849	0.835	0.828	0.805
12	1.92	1.14	0.975	0.902	0.865	0.840	0.811	0.784	0.771	0.758	0.752	0.732
14	1.77	1.05	0.899	0.831	0.797	0.775	0.748	0.723	0.710	0.699	0.693	0.676
16	1.65	0.976	0.838	0.775	0.743	0.722	0.697	0.673	0.662	0.651	0.646	0.631
18	1.56	0.921	0.790	0.731	0.701	0.681	0.658	0.635	0.624	0.614	0.609	0.594
20	1.48	0.873	0.750	0.693	0.665	0.646	0.624	0.602	0.592	0.583	0.578	0.563
25	1.32	0.779	0.669	0.619	0.593	0.577	0.557	0.538	0.529	0.520	0.515	0.502
30	1.20	0.708	0.608	0.563	0.540	0.525	0.507	0.489	0.481	0.473	0.469	0.456
40	1.04	0.613	0.526	0.486	0.467	0.454	0.438	0.423	0.416	0.409	0.405	0.395
50	0.925	0.548	0.471	0.435	0.417	0.405	0.391	0.378	0.371	0.365	0.362	0.353
60	0.844	0.499	0.429	0.396	0.380	0.369	0.356	0.344	0.338	0.333	0.330	0.322
80	0.730	0.432	0.371	0.342	0.328	0.319	0.308	0.298	0.292	0.288	0.285	0.278
100	0.652	0.385	0.331	0.306	0.293	0.285	0.275	0.266	0.261	0.257	0.255	0.249

FIG. 2. Determination of second sample size.* Values of a/s_1 for $\alpha = 0.05$ for one-sided tests, $\alpha = 0.10$ for two-sided tests $\beta = 0.20$.

* Reproduced with permission from M. HARRIS, D. G. HORVITZ, A. M. MOOD, On the determination of sample sizes in designing experiments. *J. Amer. Statist. Ass.* **43,** 391 (1948).

levels in vegetation. Also, the activity in vegetation fluctuates with the season as stated by HORTON.[2]

Following are the computations of the power of the test or $1 - \beta$ values for sample sizes $N = 24$ and $N = 16$. Assume

$$s = 68.7$$

$$H: \ \mu = \mu_0 = 60$$

It is desirable to detect $\mu = 100$ when $\alpha = 0.05$. To determine the power, compute the following:

$$\frac{\mu - \mu_0}{s/\sqrt{N}}$$

For $N = 24$

$$\frac{100 - 60}{68.7/\sqrt{24}} = 2.85$$

From the graph shown in Fig. 3 on the horizontal scale, it is found 2.85 corresponds to a power 0.90 on the vertical scale, using the *a* curve. This means that if the true μ is actually 100 there is a chance $1 - \beta = 0.90$ of rejecting the $H: \ \mu = \mu_0 = 60$ and it is concluded that there is a significant increase in activity. For $N = 16$ it is found:

$$\frac{100 - 60}{68.7/\sqrt{16}} = 2.33$$

This value gives a power of 0.78. Thus, for silt samples this means if the true μ is actually 100, there is a chance $1 - \beta = 0.78$ of rejecting the $H: \ \mu = \mu_0 = 60$, and it is concluded there is a significant increase in activity. The value of $1 - \beta = 0.78$ is a somewhat low value for the power of the test. In view of the fact that less emphasis is placed on silt samples than on water, soil and vegetation, it is considered that the value of $1 - \beta = 0.78$ is sufficient.

Incidents

If an incident is known to occur, a complete set of samples are taken within 48 hr. These samples are processed and analyzed as soon as possible. A separate report is filed on the findings and the data is not made an integral part of the report of routine samples.

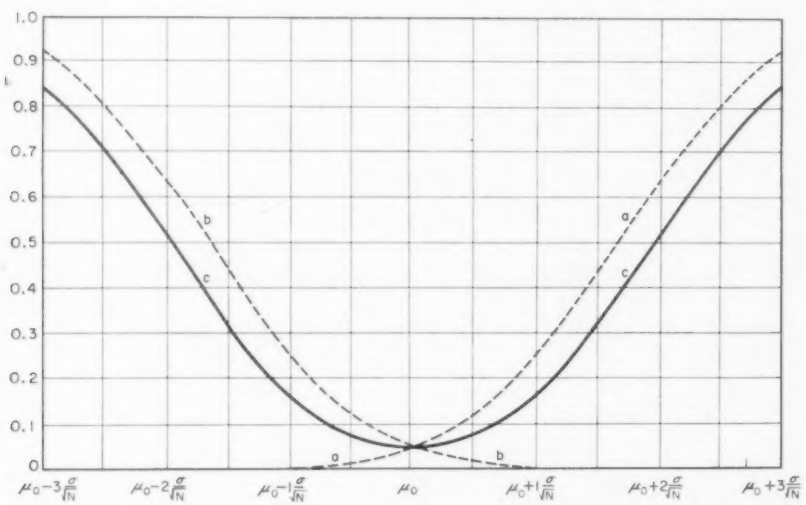


FIG. 3. Statistical graph. Power functions for three tests of hypothesis: $\mu = \mu_0$ in normal distribution, $\alpha = 0.05$. Let $z = \dfrac{X - \mu_0}{\sigma/\sqrt{N}}$; (a) reject if $z > 1.645$; (b) reject if $z < -1.645$; (c) reject if $z > 1.96$ or if $z < -1.96$. (Reproduced from *Introduction to Statistical Analysis* by W. DIXON and F. MASSEY, by courtesy of the McGraw-Hill Book Company, Inc.)

E. SAMPLING

1. *Water*

As determined above, water samples are taken twice monthly. These samples are taken in all annuli except 1 and 2. There are very few water sources in these annuli and no ponds. In all sectors of the remaining annuli except 3.1 and 4.4 water samples are taken from ponds.

The absence of water samples in annuli 1 and 2 is partially compensated for by a separate river sampling program and a sampling program for well water on the site. Procedures used in preparing samples are similar to those of several major laboratories and the activity determined by counting room personnel. Wells supplying drinking water for site personnel are sampled twice monthly.

2. *Soil*

Soil samples are collected twice montly in each sector of each annulus. Each sector of an annulus is divided into nine sub-sectors for purposes of random sampling. These samples are taken from a surface area of approximately 1 ft² and within 1 in. of the surface. Soil sample preparation is accomplished by methods accepted at several other laboratories.

3. *Silt*

The method of collection and procedures for these samples are the same as for soil samples. However, two samples monthly are collected in all annuli except 1 and 2 for the months of April to November inclusive. Silt samples are unavailable in the remaining months due to the presence of ice.

4. *Vegetation*

Vegetation samples are obtained from each sector of each annulus twice monthly. Each sector of an annulus is divided into nine sub-sectors for purposes of random sampling. Collection of samples is limited to grass due to its ubiquitous nature. Also, results on one species of plant are more significant than comparisons of a variety of plants.

Samples are collected from an area approximately 1 ft². Commonly accepted sample preparation practices are followed.

5. *Air*

Daily air samples are taken with a Staplex Hi-Volume air sampler at a central location on site. In addition, frequent air samples are taken at locations both on and off the CANEL site. These samples are held for seven days to allow for decay of radon and thoron daughters before counting.

F. TREATMENT AND ANALYSIS OF DATA

The counting room reports bear all necessary data for statistical computations. The figures of $\mu\mu$c/g or $\mu\mu$c/l. are entered on the form in Fig. 4. This table is the Master Table necessary for performing the three-way anlaysis of variance. One of these tables is necessary for each category, i.e. soil–α, soil–β, water–α, etc. This method of analysis is outlined by KENDALL.[3] It shall suffice here to say that the method is extremely sensitive, and a great deal of confidence may be placed in the results of it.

Date	Direction												Total
	South				West				North				
	Distance				Distance				Distance				
	A	B	C	Total	A	B	C	Total	A	B	C	Total	
Spring	80* / +30	95 / +45	83 / +33	258 / +108	53 / +3	42 / −8	48 / −2	143 / −7	51 / +1	43 / −7	53 / +3	147 / −3	548 / +98
Summer	41 / −9	48 / −2	55 / +5	144 / −6	50 / 0	53 / +3	49 / −1	152 / +2	60 / +10	41 / −9	47 / −3	148 / −2	444 / −6
Fall	54 / +4	39 / −11	53 / +3	146 / −4	44 / −6	48 / −2	49 / −1	141 / −9	50 / 0	55 / +5	43 / −7	148 / −2	435 / −15
Total	175 / +25	182 / +32	191 / +41	548 / +98	147 / −3	143 / −7	146 / −4	436 / −14	161 / +11	139 / −11	143 / −7	443 / −7	1427 / +77

FIG. 4. Master table—three-way analysis of variance.* $N = 27$; assume $x = 50$.

* To obtain value in $\mu\mu$c/g or l, multiply by 10^{-2}.

A few comments may be made about the items of Fig. 4 and the manner in which they are entered. The table shown is merely an example and the numbers within this table are artificial. There is no limit to the number of dates, directions or distances which may be used. Data occurring as whole numbers are not normally found. For computational purposes, it is convenient to have whole numbers. If one has numbers appearing as follows: 45.26, 36.81, 41.3, 35.4, 40.8 . . . , the decimal is merely moved one place to the right and rounded off to the nearest whole number. Moving of the decimal is noted in Fig. 4. The above numbers would then appear as 453, 368, 413, 354, 408 . . . These numbers are entered to the left of the slash in Fig. 4. The figures needed for further computations will be in the form of deviations from some mean. This does not have to be a true mean but may be an assumed mean obtained by glancing at the data. The assumed $\bar{x}$ in the example was 50. Deviations from this assumed mean with plus or minus sign are entered to the right of the slash. One important fact which cannot be overlooked is that every box in Fig. 4 must be filled. The remainder of the analysis is performed by standard three-way analysis of variance techniques.

This analysis can be readily adapted to machine computation and is at present being performed on an IBM 704.

The analysis of variance procedure is designed to detect significant changes. It is of secondary interest to separate contamination due to weapons testing from contamination due to CANEL operations.

If the analysis of variance were to be performed after every sample period, it would involve a considerable amount of time. Therefore, the analysis will be performed at intervals of 3 months, 6 months, 9 months, and 1 year. After a 1-year cycle has been completed, the analysis shall be performed every 3 months using the previous 12-months data. This prevents the master table from becoming unwieldy due to its extension past one year's data. This technique fulfills objective (1) as stated in Section B.

For well water and air samples, t-tests are used for comparison with MPC's. This is done as follows. Hypothesis: Universe mean is not greater than a specific number when σ^2 is not known (however, σ^2 is estimated by S^2).

(a) H: $\mu \leq \mu_0$ and $\mu_0 = \text{MPC}$;

(b) let $\alpha = 0.05$;

(c) use $t = \dfrac{\bar{x} - \mu_0}{S/\sqrt{N}}$ to test the hypothesis;

(d) assuming that the universe has a normal distribution, then this statistic has a $t(N-1)$ distribution with percentiles recorded in Fig. 5.

(e) the critical region is $t > t_{1-\alpha}$; it is desired to reject the hypothesis only when t is large, say larger than $t_{1-\alpha}$, which gives the chosen level of significance α;

(f) compute the value of t and reject the hypothesis if the observed t is greater than or equal to $t_{1-\alpha}$ or accept the hypothesis if the observed t is less than $t_{1-\alpha}$.

If it is desired to compare some portion of current data with similar portions of data for some past period, it is accomplished by use of another t-test as follows. Hypothesis: two populations have the same mean when σ^2 is not known.

(a) H: $\mu_1 = \mu_2$; assume $\sigma_1^2 = \sigma_2^2 = \sigma^2$ but σ^2 not known;

(b) let $\alpha = 0.05$;

(c) to test the hypothesis, use the following statistic:

$$t = \frac{\bar{x}_1 - \bar{x}_2}{S_p\sqrt{(1/N_1 + 1/N_2)}}$$

where S_p^2 is the pooled mean-square estimate of σ^2 computed by

$$S_p^2 = \frac{(N_1 - 1)S_1^2 + (N_2 - 1)S_2^2}{N_1 + N_2 - 2}$$

(d) If both populations have normal distributions with the same mean and the same variance, then this statistic has the $t(N_1 + N_2 - 2)$ distribution with percentiles recorded in Fig. 5.

(e) The rejection region is $t < t_{\alpha/2}(N_1 + N_2 - 2)$ and $t > t_{1-\alpha/2}(N_1 + N_2 - 2)$

(f) Compute t and reject or accept the hypothesis.

d.f.	$t_{0.95}$	$t_{0.975}$	$t_{0.9875}$	$t_{0.995}$	$t_{0.9975}$
1	6.31	12.7	25.5	63.7	127
2	2.92	4.30	6.21	9.92	14.1
3	2.35	3.18	4.18	5.84	7.45
4	2.13	2.78	3.50	4.60	5.60
5	2.01	2.57	3.16	4.03	4.77
6	1.94	2.45	2.97	3.71	4.32
7	1.89	2.36	2.84	3.50	4.03
8	1.86	2.31	2.75	3.36	3.83
9	1.83	2.26	2.69	3.25	3.69
10	1.81	2.23	2.63	3.17	3.58
11	1.80	2.20	2.59	3.11	3.50
12	1.78	2.18	2.56	3.05	3.43
13	1.77	2.16	2.53	3.01	3.37
14	1.76	2.14	2.51	2.98	3.33
15	1.75	2.13	2.49	2.95	3.29
16	1.75	2.12	2.47	2.92	3.25
17	1.74	2.11	2.46	2.90	3.22
18	1.73	2.10	2.45	2.88	3.20
19	1.73	2.09	2.43	2.86	3.17
20	1.72	2.09	2.42	2.85	3.15
21	1.72	2.08	2.41	2.83	3.14
22	1.72	2.07	2.41	2.82	3.12
23	1.71	2.07	2.40	2.81	3.10
24	1.71	2.06	2.39	2.80	3.09
25	1.71	2.06	2.38	2.79	3.08
26	1.71	2.06	2.38	2.78	3.07
27	1.70	2.05	2.37	2.77	3.06
28	1.70	2.05	2.37	2.76	3.05
29	1.70	2.05	2.36	2.76	3.04
30	1.70	2.04	2.36	2.75	3.03
40	1.68	2.02	2.33	2.70	2.97
60	1.67	2.00	2.30	2.66	2.91
120	1.66	1.98	2.27	2.62	2.86
∞	1.64	1.96	2.24	2.58	2.81
d.f.	$t_{0.05}$	$t_{0.025}$	$t_{0.0125}$	$t_{0.005}$	$t_{0.0025}$

When the table is read from the foot, the tabled values are to be prefixed with a negative sign. Interpolation should be performed using the reciprocals of the degrees of freedom. The values in the above table were computed from percentiles of the *F* distribution.

FIG. 5. Values of t*

* Reproduced from *Introduction to Statistical Analysis*. By W. Dixon and F. Massey, by courtesy of the McGraw-Hill Book Company, Inc.

Table 1. *Multipliers for control charts**

N	D_L	D_U	A_2
2	0.009	3.52	1.88
3	0.100	2.58	1.02
4	0.185	2.26	0.73
5	0.254	2.09	0.58
6	0.308	1.97	0.48
7	0.351	1.90	0.42
8	0.386	1.84	0.37
9	0.415	1.79	0.34
10	0.441	1.76	0.31
11	0.463	1.72	0.29
12	0.482	1.70	0.27
13	0.498	1.68	0.25
14	0.511	1.66	0.24
15	0.524	1.64	0.22

* Reproduced from *Introduction to Statistical Analysis by* DIXON, MASSEY, by courtesy of the McGraw-Hill Book Co. Inc.

To meet the requirements of objective (2), control charts on means and ranges will be used. The question might be asked, "why a control chart on both means and ranges?" Take the numbers 20, 50, 80, 110 for example and find a mean of 65 which may be close to a desired mean, but the range from 20 to 110 may be entirely too large. A mean μ for some basic data which may consist of as little as four or five sampling periods can be established. The charts can readily be recomputed whenever desired, and it is expected that the control limits would narrow each time. This method of comparison needs to be very rapid to give a running analysis of the data. This can be accomplished by using the method of DIXON and MASSEY[4]. It is explained briefly here with an example shown.

Assume there is past data for 12 months or twenty-four sampling periods. Each sampling period there are twenty water samples, for example. Compute a mean μ for these 480 samples. Compute a range R for each set of twenty samples and a mean range

$$\bar{R} = \frac{\Sigma R}{24}$$

where $R = X$ maximum $- X$ minimum. Assume there is data where $\mu = 50$ and $\bar{R} = 48$. These are not unreasonable figures for water data although they are not actual figures. For this case $N = 24$ and Table 1 is entered with $N = 15$. An extension of this table to higher values of N is not available at the present time, but $N = 15$ is sufficient for this example. From the table it is readily seen that D_L, D_U and A_2 are not decreasing rapidly at $N = 15$.

To compute control limits on μ, use $\mu \pm A_2\bar{R}$ and for this example is:

$$50 \pm (0.22)(48) = 50 \pm 10.56 = 39.4 \text{ to } 60.6$$

These are 3σ limits and the chart is shown in Fig. 6. Thus, when a set of samples is taken, merely plot the $\bar{x}$ on this chart.

Next the limits for $\bar{R}$ are plotted. These limits will be at 2.576σ. Using $\bar{R} = 48$ and the following relation for limits one finds:

$$D_L\bar{R} \text{ to } D_U\bar{R} \text{ with } N = 15;\ (0.524)(48) \text{ to } (1.64)(48) = 25.2 \text{ to } 78.7$$

Thus, every time a set of samples are taken, the R is computed as $X_{\max} - X_{\min}$ and plotted on the range chart shown in Fig. 7.

The control charts are computed for a maximum of 1 year's data. Previous to that, they are computed at the end of 3, 6, 9 months, respectively.

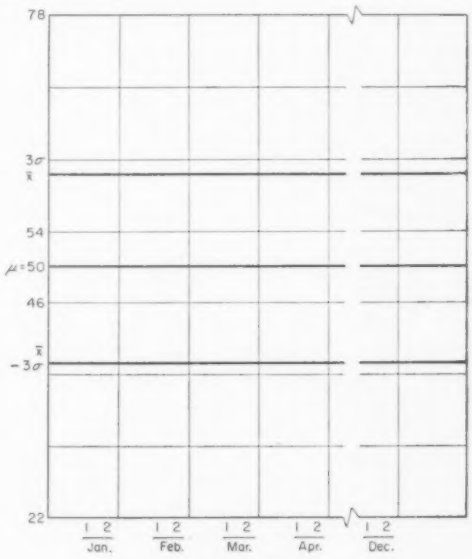


FIG. 6. $\bar{x}$-Chart.

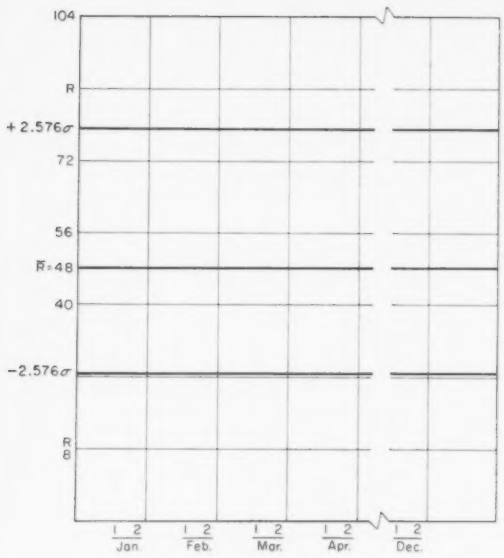


FIG. 7. Range chart.

The fulfillment of objective (3) is a simple procedure. Annual averages of air and water concentrations are computed and statistically compared with MPCs.

G. INTERPRETATION OF DATA

Whenever a sample or set of samples shows a significant increase in activity, it will be a matter of concern for that one sample or set of samples. Two occasions of significant increase in activity may or may not be treated alike. Each event may present its own peculiar aspects. Therefore, for many cases, no set pattern may be established for the investigation of the results.

The analysis of variance procedure mentioned in Section F is an extremely sensitive method of analysis. Using the three variables of classification, i.e. direction, distance and date, the following effects can be found:

(1) Direction effect.
(2) Distance effect.
(3) Date effect.
(4) Direction–distance effect.
(5) Direction–date effect.
(6) Distance–date effect.
(7) Direction–distance–date effect.

If it is found that direction is an influencing factor, the basic data can then be re-examined to localize which direction or directions are responsible. If, at the same time, it is found that distance is an influencing factor, the basic data could be examined to find which distance or annulus is responsible. If the above conditions were true, it follows that a first-order interaction of direction and distance would be significant. Thus, the analysis of variance technique is very specific in telling the effect of the three variables. The above techniques apply to soil, silt, water and vegetation samples. Precipitation samples may be analyzed by analysis of variance techniques, but it cannot be a three-way analysis. These samples will be analyzed using wind direction and time as variables of classification, but they must be used separately using one variable at a time.

Well-water samples are treated more simply. They are not classified by date, direction and distance. A straight comparison of current samples with base data is made using the second t-test as described in Section F. A comparison of yearly average concentrations with the MPC of 100 $\mu\mu$c/l. is made, using the first t-test described in Section F. Air samples and other water samples may be treated similarly as well-water samples. Comparison of annual average concentrations with MPC's are made. Information which is helpful in localizing significant increases in activity is the result of analysis of variance techniques, meteorological data and studies of facility operations for the period of time concerned.

The use of control charts on means and ranges is extremely valuable in quickly detecting trends of activity increase or of increases in one sampling period. For example, if it is found in a set of twenty-eight samples readings ranging from 10 to 100 and an $\bar{x}$ of 50, nothing would be noted on the $\bar{x}$-chart, but lack of control would be indicated on the R-chart. This would be cause for an investigation or at least confirmation of the sample. The alternate situation might exist if samples are found ranging from 55 to 85 with $R = 30$ but $\bar{x} = 77$. Nothing would be noted on the R-chart, but a significant increase would be indicated on the $\bar{x}$-chart.

Actually, the values which fall below the $\overline{R}$

or μ on the control charts are of little significance but might indicate instrument trouble or some similar factor. The values above the R and μ are the ones of great significance.

Fallout data is plotted on a graph showing dis/min per ft^2 vs. date. This data is correlated with information on AEC tests, Russian tests, etc.

At some date in the future, it may be possible to establish correlation coefficients between, for example, water samples taken at specific locations and soil samples taken at random within a sector of an annulus. This is merely anticipation at the present time but some attempt to do so shall be made. If the correlation coefficient could be established, it would then be possible to abandon either soil or water samples. Statistical procedures alone are not the basis for interpretation of data. Interpretation is made upon the basis of statistical analyses, meteorological data, facility operations and stack monitor data. Statistical analyses, however, give the first indication of significant increase in activity.

REFERENCES

1. NBS Handbook 52.
2. J. H. Horton, E. I. DuPont Co., Savannah River Plant, Aiken, South Carolina; to Dr. G. B. Wheeler, personal communication of 2 February 1956.
3. M. G. Kendall, *Advanced Theory of Statistics*, Vol. II. Driggin and Co., Ltd., London (1948).
4. W. Dixon and F. Massey, *Introduction to Statistical Analysis*. McGraw-Hill, New York (1951).

Health Physics Pergamon Press 1961. Vol. 6, pp. 163–176. Printed in Northern Ireland

THE EFFECT OF WHOLE BODY X-IRRADIATION ON THE BLOOD AND OTHER TISSUES OF WHITE LEGHORN COCKERELS

R. L. ATKINSON, J. H. QUISENBERRY, A. B. MEDLEN and G. BERGER*
Department of Poultry Science, Agricultural and Mechanical College of Texas, College Station, Texas

(*Received* 2 *February* 1961)

Abstract—White Leghorn cockerels were given levels of whole body X-irradiation ranging from 50 to 500 r at 6, 12 and 18 weeks of age. The irradiation was given at 160 kV, 10 mA, and with a target distance of 20 in. Half a millimeter of copper and 1 mm of aluminum were used to filter the rays which were delivered to the target at a rate of approximately 16 r/min.

Blood samples were taken at 1, 3, 8 and 18 days after irradiation and were used to determine total red cell count, total white cell count, hemoglobin and white cell differentials. Irradiated birds not used for blood counts and hemoglobin determinations were sacrificed at 1, 3 and 5 weeks after irradiation in order to study the effects of irradiation upon certain tissues.

All of the blood data indicate that in general the cockerel reacts to X-irradiation in a manner similar to other animals. The large number of basophils in the peripheral blood make the animal an excellent subject for a study of the effect of irradiation on this particular cellular element.

Tissue damage was found to be similar to that reported for other animals. Irradiation damage was manifested in the seminiferous tubules of the testes as follows: (1) sperm scattered among the cells comprising the tubular walls; (2) a decrease in the number of sperm attached to Sertoli cells; (3) sperm oriented in an abnormal manner; (4) the presence of cell necrosis; and (5) a massive sloughing of cells in the tubules.

I. INTRODUCTION

THERE has been a great deal of interest in the effect of X-irradiation on the blood and other tissues of animals. The literature prior to 1939 has been reviewed by BIGNOMI[1]. MULLIGAN[2] reported that, in dogs, repeated small doses of roentgen rays caused a depression of the total leucocyte count and an occasional eosinophilia, resulting in a moderate anemia in the peripheral blood. Similar results have been reported by other workers.[3–6] A lymphopenia following whole body X-irradiation of mice was noted by DOUGHERTY and WHITE[7]. These workers suggested that the X-rays exerted both a direct and an indirect effect on the lymphocytes. The effect of X-rays on the lymphocytes has also been reported by JACOBSON *et al.*[8]. These workers stated that the most sensitive indicator of radiation effects in any animal that could be recognized by a study of the peripheral blood was a reduction in the number of lymphocytes. Other workers[9–11] have reported similar results for various types of animals. A slight depression was noted in the red cell count and hemoglobin content of the peripheral blood of dogs subjected to 500 r whole-body X-ray treatment.[11] LUCAS and DENINGTON[12] studied the effect of whole-body X-irradiation on the peripheral blood of White Leghorn females and reported a severe destruction of all cellular elements.

STRANGEWAYS and HOPWOOD[13] were among the first to recognize the effects of X-rays on growing cells. These workers reported an inhibition of the onset of mitotic division.

* Present address: Crescent Park Veterinary Hospital, 1813 E. 8th St., Odessa, Texas.

ROSENBAUM[14] irradiated the pancreas of chicks and reported a destructive action along with a stimulation of the regenerative process. Other workers[4–6,15–16] have studied this destructive effect of X-rays on animal cells. The cells most sensitive to irradiation were reported to be the lymphocytes, gonadal cells and epithelial cells.

A large number of workers[17–28] have studied the effects of whole-body X-irradiation or the application of X-rays directly to the testes. It has been reported that, depending upon the type and length of treatment, the following effects have been observed: (1) degeneration of spermatogonia; (2) depopulation of the seminal canal; (3) the formation of abnormal giant cells, both mononuclear and multinuclear; and (4) destruction of the interstitial cells.

The present study was undertaken to study more fully the effects of whole-body X-irradiation on a number of tissues, and to determine if the chick would respond to irradiation in a manner similar to other experimental animals.

2. METHODS

Groups of White Leghorn cockerels were given whole body X-irradiation at 6, 12 or 18 weeks of age as shown in Table 1. All animals were irradiated with a 200-kV Westinghouse Quadro–Condex Deep Therapy X-ray unit. The irradiation was given at 160 kV, 10 mA, and with a target distance of 20 in. Half a millimeter of copper and 1 mm of aluminum were used to filter the rays which were delivered to the target at a rate of approximately 16 r/min as measured by a Victoreen dosimeter. Each bird received half of the dose on one side of the body and was turned over and given the other half of the dose on the opposite side. Birds were constrained between layers of cheesecloth to prevent movement.

Five birds from each group were used for a study of the cells of the peripheral blood. Five different birds from each irradiation group were used for hemoglobin determinations where sufficient numbers were available.

Blood samples were obtained at 1, 3, 8 and 18 days after irradiation. In the group which was treated with two doses of 50 r, 1 week apart, the samples were taken only after the second irradiation. The blood samples were obtained by puncturing the median vein of the wing at the humeroradial articulation with a sharp, twenty-gauge hypodermic needle. The first drop of blood to flow from the vein was omitted from the collection, after which samples for cell counts, blood smears and hemoglobin determinations were taken.

Table 1. *Effect of age and various levels of whole body irradiation on post-irradiation mortality of White Leghorn cockerels*

Age when irradiated	X-ray dosage in r units	Number of birds irradiated	% mortality 30 days after irradiation
6 weeks	0	14	0
	50	14	7.1
	50 + 50*	14	0
	100	14	0
	200	14	0
	300	14	7.1
	500	14	21.4
12 weeks	0	11	0
	50	11	0
	50 + 50*	11	9.1
	100	11	0
	200	11	0
	300	11	9.1
	500	11	54.5
18 weeks	0	9	0
	50	9	11.2
	50 + 50*	9	11.2
	100	9	0
	200	9	0
	300	9	11.2
	500	7	42.9

* Two treatments of 50 r each given 1 week apart.

Hemoglobin was determined by the direct photometric method (HAWK *et al*[29]), in which the blood was diluted in a weak alkaline solution and hemoglobin determined by colorimetric measurement.

The red cell and white cell counts were made according to the method of NATT and HERRICK[30]. A Trenner automatic red cell pipette was used in making the dilution (1 to 100) and cell counts were made with a Spencer "Bright-Line" hemocytometer and were counted in the normal manner of counting both erythrocytes and leucocytes. Where there did not appear to be a normal dispersion of the cells, duplicate counts were made.

The blood smears were stained with Wright's stain, and the percentage of each of the different types of leucocytes was obtained. It was found that the addition of 1 minim of acetic acid to each 250 ml of

the diluted stain would give much better staining qualities. To determine the differential percentage of the leucocytes, 200 leucocytes were counted and the percentage of each cell type determined. The absolute value was obtained by multiplying the percentage of each cell type by the total leucocyte count.

Irradiated birds not used for blood counts and hemoglobin determinations were sacrificed at 1, 3 and 5 weeks after irradiation. The birds were opened as quickly as possible and tissue samples taken.

The testes samples were fixed in Bouin–Allen fixative, dehydrated and cleared in alcohol and cedar wood oil, imbedded in tissuemat, sectioned at 5 μ and stained with Heidenhain's iron hematoxylin. Samples of spleen, thyroid and pancreas were fixed in both Bouin–Allen fixative and neutral formalin (10 per cent). The same procedure for dehydrating, clearing, imbedding and sectioning was followed as described for the testes samples. The spleen and thyroid sections were stained with Weigert's hematoxylin–eosin, while the pancreas sections were stained with Harris's hematoxylin–eosin. The pituitaries were fixed in Zenker–formol solution and were dehydrated, cleared, imbedded and sectioned as described above for the other tissues. The pituitary sections were stained by the Mallory–Azan method. This is a modification of the Mallory triple stain in which azo-carmine is substituted for stain 1 of the Mallory triple stain. In this technique the connective tissue stains blue as well as the few β-cells. The two types of α-cells stain a deep red or an orange. Red blood cells stain a deep red.

3. RESULTS AND DISCUSSION

Effect of irradiation on the peripheral blood

The lymphocytes. The lymphocytes showed extreme damage with considerable deviation from the normal (Table 2). The greatest decrease was observed in birds irradiated with a whole-body dosage of 500 r. The period in which the lowest number of lymphocytes was observed was found to be during the first day following irradiation. This was generally true for all age groups and for all levels of irradiation. From this low level there seemed to be a gradual recovery phase for all treatment groups. The rate of recovery appeared to be related to age with the younger birds recovering at a more rapid rate. These results are similar to that reported by other workers[4–12] for other animals with the exception of one report[9] that in guinea-pigs the lowest level occurred around the ninth day after irradiation.

The heterophilic count. X-irradiation caused a slight increase in the heterophilic count (Table 3) during the first day following treatment. This

Table 2. Effect of roentgen-rays on the lymphocytes

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	8753	8805	6278	2831	1015	354	14,548
	3	9083	8141	7688	5302	3968	2520	11,424
	8	9383	9880	8422	7560	5777	4712	16,426
	18	13,028	8832	9095	8758	9623	11,204	13,910
12 weeks	1	8257	7479	7189	3976	2030	348	15,382
	3	11,005	8392	9881	7142	2640	1746	12,456
	8	10,991	11,558	7018	10,098	5416	1696	11,946
	18	10,836	10,148	9840	8369	8577	6761	13,606
18 weeks	1	8781	7819	6010	3495	1443	334	14,410
	3	8568	8438	7283	4274	2195	1812	13,151
	8	9819	7405	9753	5135	4482	2591	15,569
	18	9412	10,890	9825	8060	5624	5676	15,980

* Two treatments of 50 r each given 1 week apart.

Table 3. Effect of roentgen-rays on the heterophils

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	4180	5601	5103	3619	4564	3563	4395
	3	2538	2691	3251	3352	3193	3102	3461
	8	2853	1846	2995	2236	598	133	3646
	18	2924	2586	1777	2192	1445	1661	2484
12 weeks	1	4133	5591	5925	5511	5788	6344	4224
	3	3994	4174	7000	4496	4163	4675	3903
	8	4744	3784	4448	5151	551	83	3825
	18	3780	3710	4412	2860	2086	1637	4431
18 weeks	1	6426	5903	7263	5405	4284	6163	5697
	3	4843	4675	6673	4809	2901	2050	5163
	8	4572	2989	5611	2430	3030	42	5495
	18	5006	3487	4361	3016	2725	1308	5194

* Two treatments of 50 r each given 1 week apart.

slight increase, however, did not appear to have any definite relationship to age or to the amount of X-rays given. The stimulation was followed by an extreme depression about the eighth day post-irradiation which appeared to be related to dosage but not age and which was followed by a gradual return toward the normal count. This recovery rate appeared to be related to both age and dosage level since the older birds, particularly at the lower dosage levels, appeared to recover more quickly. At higher dosage levels, recovery was very slight at 18 days after exposure regardless of the age of the bird. It would appear that the effect of X-irradiation on the heterophilic count is characterized by a slight stimulation immediately following treatment, followed by a depression at 8 days post-irradiation with a gradual return to the normal level with the effects being somewhat related to both age and dosage level. These results are similar to those reported by other workers[2,8–10,12] with the exception that in one case[9] the initial stimulation was not observed and in another[12] there was reported a definite relationship between the age of the animal and the initial stimulatory effect.

The monocytic count. The effect of X-irradiation on the monocytic count (Table 4) was found to be quite similar to that noted for the lymphocytic count. The general effect was to cause a severe depression, particularly at the higher dosage levels, at about the eighth day after irradiation followed by a gradual return to the normal level. This return to normal was somewhat faster than that observed for the lymphocytes. Other workers[8,9,12] have reported results which are quite similar.

The basophilic count. Results indicated that the basophils were extremely sensitive to destruction by X-rays. Birds irradiated at 6 weeks of age made a very fast recovery, being essentially normal by the eighteenth day following treatment. A rather slow recovery was noted in birds irradiated at 12 or 18 weeks of age, particularly at the higher dosage levels (Table 5). The most severe drop was noted in birds given 500 r of whole body X-irradiation. This drop was from the normal (500–600) to zero. Four-hundred cells were counted to determine the differential percentage in each case where no basophils were found. In general, the recovery phase started about the eighth day after irradiation, and as in the lymphocytic and monocytic counts the rate of recovery was dependent upon the age of the birds and the size of the irradiation dose but was much slower than that noted for the lymphocytes and monocytes. The cockerel appears to be a very good

Table 4. Effect of roentgen-rays on the monocytes

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	1642	1191	1715	1193	1134	531	1550
	3	1540	1147	1484	872	395	312	1201
	8	911	1008	973	799	401	440	2260
	18	1564	1088	1348	1273	1290	1570	1362
12 weeks	1	1496	1073	2228	659	679	845	1833
	3	1686	1465	1963	1492	559	358	1537
	8	1539	1359	844	1411	748	212	1799
	18	1344	1038	1460	1398	1281	763	1495
18 weeks	1	1318	1460	1359	832	479	405	1522
	3	1028	1058	1790	1071	403	302	2050
	8	1794	1415	1841	997	713	261	2314
	18	2031	1650	1515	1482	1155	837	1457

* Two treatments of 50 r each given 1 week apart.

Table 5. Effect of roentgen-rays on the basophils

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	684	368	338	62	7	0	708
	3	630	298	374	127	54	0	605
	8	243	176	346	173	17	16	471
	18	508	230	365	302	490	393	438
12 weeks	1	329	72	269	41	0	0	727
	3	932	562	364	243	75	7	676
	8	537	464	227	323	78	8	659
	18	806	346	590	342	195	140	545
18 weeks	1	374	361	272	59	6	0	471
	3	253	456	488	242	56	0	363
	8	505	406	389	125	66	6	627
	18	400	393	594	351	262	63	729

* Two treatments of 50 r each given 1 week apart.

animal in which to study the basophils due to the large number of these cells found in the peripheral blood. The results are different from those reported[9] for the guinea-pig but are identical to those reported[12] for the White Leghorn female.

The eosinophilic count. The eosinophilic pattern (Table 6) showed a definite initial stimulatory effect following 500 r whole-body X-irradiation. This initial stimulatory effect (24 hr) was followed by a sharp reduction (to zero in case of the 500 r groups). The eosinophilic count had returned to normal, in birds irradiated at 6 weeks of age, by the eighteenth day after

Table 6. Effect of roentgen-rays on the eosinophils

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	122	191	81	78	210	346	173
	3	210	124	103	147	140	66	118
	8	119	91	64	32	7	0	46
	18	114	64	50	76	52	272	138
12 weeks	1	37	86	190	113	103	363	253
	3	133	207	171	128	64	89	257
	8	90	34	63	17	7	1	73
	18	34	135	98	33	61	0	178
18 weeks	1	85	157	196	109	88	199	29
	3	209	74	41	105	45	36	27
	8	140	86	106	36	8	0	96
	18	285	83	82	91	44	16	212

* Two treatments of 50 r each given 1 week apart.

Table 7. Effect of roentgen-rays on the total leucocytes

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	15,200	14,700	13,500	7800	7000	4700	21,375
	3	14,000	12,400	12,900	9800	7750	6000	16,800
	8	13,000	13,000	12,800	10,800	6800	5300	22,875
	18	18,125	12,800	12,600	12,600	12,900	15,100	18,400
12 weeks	1	14,250	14,300	15,800	10,300	8600	7900	22,375
	3	17,750	14,800	19,375	13,500	7500	6875	18,625
	8	17,900	17,200	12,600	17,000	6800	2000	18,300
	18	16,800	15,375	16,400	13,000	12,200	9300	20,250
18 weeks	1	17,000	15,700	15,100	9900	6300	7100	22,125
	3	14,900	14,700	16,275	10,500	5600	4200	20,750
	8	16,833	12,300	17,700	8900	8300	2900	24,100
	18	17,166	16,500	16,375	13,000	10,900	7900	23,500

* Two treatments of 50 r each given 1 week apart.

treatment. Recovery had not even begun in the older birds and particularly those irradiated at 12 weeks of age. In general, X-irradiation appeared to cause an immediate stimulation (at least following treatment of 500 r) followed by a drastic reduction and a delayed recovery, particularly in the older birds. These data are in conflict with those reported by other workers[9,12] who failed to note any initial stimulatory effect following irradiation.

The total leucocyte count. The total leucocyte count (Table 7) resembles that seen for the lymphocyte, since the lymphocytes make up approximately 70 per cent of the total leucocytes. There was a definite drop in number the first day after treatment, which was the lowest

Table 8. Effect of roentgen-rays on the erythrocytes

Age at irradiation	Days after irradiation	Number of cells in each cubic millimeter of blood*						
		50 r	50 + 50 r†	100 r	200 r	300 r	500 r	Control
6 weeks	1	2.41	2.50	2.55	2.68	2.68	2.83	2.80
	3	2.36	2.69	2.45	2.90	2.61	2.78	2.65
	8	2.51	2.74	2.70	2.54	2.49	2.36	2.93
	18	2.72	2.71	3.06	2.86	2.83	2.83	3.05
12 weeks	1	3.03	2.78	3.10	3.20	3.05	3.08	2.82
	3	2.94	2.94	2.94	3.22	2.72	3.00	2.79
	8	3.05	2.95	2.86	3.02	2.68	2.71	2.82
	18	2.80	2.87	3.01	2.97	2.54	2.51	2.71
18 weeks	1	2.97	2.98	3.09	3.22	3.31	3.27	2.79
	3	2.84	2.95	2.75	3.09	3.34	3.29	3.01
	8	3.20	2.89	3.08	2.19	3.21	3.10	3.08
	18	3.00	3.36	3.02	3.24	3.21	3.09	3.09

* Expressed in millions.
† Two treatments of 50 r each given 1 week apart.

point with regard to birds irradiated at 6 weeks of age. The lowest point was noted 8 days after irradiation in the older birds, particularly those given 500 r whole-body X-irradiation. The recovery phase began on approximately the eighth day after treatment. The rate of recovery, in general, decreased with increased age and with the irradiation level. These data, in general, are similar to those reported by other workers[4–11] for other animals in which this initial drop and gradual recovery to the normal level have been noted and to that reported[12] for the White Leghhorn female.

Erythrocyte count and hemoglobin level. There was some evidence (Tables 8 and 9) for a slight stimulatory effect on the erythrocyte count and hemoglobin level, followed by a return to normal. Age appeared to be a factor as well as dosage level since these irradiation effects were noted primarily in the higher (200–500 r) dosage levels and in birds either 12 or 18 weeks of age. These data are in conflict with those reported by other workers[11,12] who indicated a slight depression in both the erythrocyte count and hemoglobin level following irradiation.

All of the blood data indicate that in general the cockerel reacts to X-irradiation in a manner similar to other animals, but that differences have been found which indicate the need for more information concerning the reliability of changes in the peripheral blood as a measure of irradiation damage. These data also indicate that the cellular elements of the blood of the cockerel are sensitive to X-irradiation which makes this animal an excellent subject for study of the irradiation effects on the cellular elements of the peripheral blood.

Effect of irradiation on the body tissues

The thyroid gland. There was no cellular damage observed which could be attributed to irradiation by X-rays.

The pancreas. There was no cellular damage observed which could be attributed to X-irradiation. These results are similar to those reported by BLOOM[52].

The pituitary. No cellular damage was observed in the anterior lobe of the pituitary which could be attributed directly to X-irradiation. However, β- (basophil) cells containing vacuoles characteristic of small castrate cells were noted in cockerels irradiated with 200, 300 and 500 r dosages at 12 and 18 weeks of age.

The spleen. The exposure of cockerels to 50 r

Table 9. Effect of roentgen-rays on the hemoglobin level

Age at irradiation	Days after irradiation	Grams of hemoglobin in each 100 cm^3 of blood						
		50 r	50 + 50 r*	100 r	200 r	300 r	500 r	Control
6 weeks	1	9.48	10.23	9.77	10.33	9.88	11.27	9.49
	3	9.65	10.41	9.54	9.95	9.76	10.72	9.66
	8	10.04	9.32	9.62	10.24	9.89	9.00	10.15
	18	9.30	11.58	8.39	9.68	8.24	8.69	9.59
12 weeks	1	9.57	9.64	9.42	9.17	9.85	10.05	10.08
	3	9.67	10.13	10.10	9.39	9.15	9.15	9.20
	8	9.91	9.49	9.36	9.35	9.54	8.57	9.88
	18	10.72	9.28	9.80	9.11	8.39	9.08	9.87
18 weeks	1	10.14	9.29	10.62	11.10	11.64	11.94	9.57
	3	9.10	9.99	9.70	9.38	10.22	9.95	8.33
	8	9.28	9.73	9.89	9.77	9.50	8.32	8.75
	18	9.32	9.81	10.34	9.31	9.23	8.19	9.73

* Two treatments of 50 r each given 1 week apart.

of X-irradiation caused an increase in the number of erythrocytes in the red pulp of the spleen regardless of the age at which irradiation was given and persisted through the third week after irradiation. There was a reduction in the number of erythrocytes at the end of the fifth week. A slight increase in the number of heterophils in the red pulp was noted during the third and fifth weeks after treatment of 18-week-old birds. The lymphocytes were very sensitive to irradiation and were least in evidence the first week after irradiation. The process of recovery from the lymphocytopenia was evident during the third week after treatment with many nodules appearing normal by the fifth week after irradiation.

There was no substantial difference in the results of the 100 r dose of irradiation regardless of whether the irradiation was given in a single dose or in two equal doses of 50 r administered 1 week apart. There was a greater increase in the number of erythrocytes in the red pulp at this level of irradiation than that found at the 50-r level. This increase persisted through the fifth week after irradiation. A small number of heterophils were observed in the red pulp during the third and fifth week after irradiation in birds irradiated at 18 weeks of age. A large number of lymphocytes composing the white pulp were killed or severely damaged. A dose of 100 r caused a greater amount of damage than that seen in the 50-r group and the process of recovery had not progressed as far as that seen in the 50-r group by the end of the fifth week following irradiation.

There was a universal infiltration of the red pulp by erythrocytes in the 200, 300 and 500 r irradiated groups regardless of the age at which the birds were irradiated. An increase in the heterophils occurred during the fifth week after irradiation in birds irradiated at either 6 or 12 weeks of age which was greater for the 12-week-old group. A similar increase was noted for birds irradiated at 18 weeks of age but this occurred 2 weeks earlier. Not only did this increased number of heterophils occur during the third week but the increase far exceeded that found in younger birds given the same amount of irradiation. More lymphocytes were killed or damaged in these irradiation groups than were killed at the lower irradiation dosages. In some of the nodules, reticular elements were the only cellular elements remaining the first week after treatment. Although recovery was evident during the fifth week after treatment, it had not progressed as far in the 18-week-old birds as it

had in the 12-week-old individuals. The 12-week-old birds, in turn, had recovered less than those irradiated at 6 weeks of age.

These data indicate that the spleen of the chicken is affected by X-rays in a manner quite similar to other animals when comparison is made with the work of MURRAY[32].

The testes. The effects of whole-body X-irradiation on the testes of young cockerels (Tables 10, 11 and 12), as judged by histological appearance, was most evident the third week after irradiation. Irradiation damage was manifested in the seminiferous tubules in several ways: (1) sperm scattered among the cells composing the walls of the tubules, (2) a decrease in the number of sperm attached to the Sertoli cells, (3) sperm oriented in an abnormal manner, (4) the presence of cell necrosis, and (5) a massive sloughing of the cells in the tubules.

The amount of observable damage which occurred depended upon: (1) the quantity of irradiation, (2) the age of the bird at time of irradiation, and (3) the stage of spermatogenesis present at the time of irradiation.

Spermatogonia were the most sensitive and sperm the most resistant germ cells to irradiation. Sertoli cells and the interstitial cells of Leydig were also very resistant to irradiation. The spermatogonial cells were severely damaged or killed by irradiation, but secondary spermatocytes and later stages continued their development to form sperm unless the dosage was of sufficient strength to cause massive sloughing. Because the primitive germ cells were sensitive to irradiation there was an interval of time in which the seminiferous tubules were, in many cases, completely depleted of germ cells.

Multinuclear giant cells appeared in the lumina of the tubules of birds irradiated at 12 weeks of age. These cells appeared to be caused by a fusion of spermatids which then became associated with the Sertoli cell. The Sertoli cell cytoplasm, with its associated and fused spermatids, then separated from the remainder of the Sertoli cell and could be observed in the lumina of the seminiferous tubules. Recovery from irradiation was evident after 5 weeks in the 12-week-old birds but it had not progressed as rapidly nor approached as normal a condition as in birds treated at an earlier age.

At the higher treatment levels the seminiferous tubules of 18-week-old birds presented a somewhat different picture. Cell membranes seemed to disappear and a molten mass of syncytial material filled the seminiferous tubules. This syncytial material became necrotic and was followed by the beginning of the recovery process. Recovery from the irradiation was not complete 5 weeks after treatment. Recovery appeared to be much slower than in birds irradiated at a younger age.

These results are in general similar to those reported by other workers.[17–28] They agree with some and disagree with others, which may be attributed to the type animal used and the conditions under which the work was done.

4. SUMMARY

(1) There was a decrease in the lymphocyte count in the peripheral blood of cockerels given whole-body X-irradiation. The lowest lymphocyte number was observed during the first day following treatment. From this low level there appeared to be a gradual recovery phase for all treatment groups.

(2) X-irradiation caused a slight stimulation or increase in the heterophilic count in the peripheral blood of X-irradiated cockerels during the first day following exposure. This stimulation was followed by an extreme depression about the eighth day after irradiation followed by a gradual return toward the normal count.

(3) The effect of X-irradiation on the monocytic count was found to be quite similar to that found for the lymphocytic count. The general effect was to cause a severe depression, particularly at the higher dosage levels, at about the eighth day after irradiation, followed by a gradual return toward the normal level.

(4) The basophils were extremely sensitive to destruction by X-rays. There was, in general, an immediate and drastic reduction in the basophilic count followed by a very slow recovery rate.

(5) The eosinophils exhibited a definite initial stimulatory response, in birds given 500 r whole-body X-irradiation. This eosinophilia was followed by a sharp reduction (to an absolute zero) in case of the 500-r groups. This

Table 10. *The effect of X-irradiation on the testes of six-week-old White Leghorn cockerels*

Time after irradiation	X-Ray dosage in r units					
	50	50 + 50*	100	200	300	500
1 week	Little noticeable damage observed.	Little noticeable damage observed.	Necrotic cells observed. Little indication of lumen formation.	Necrotic cells observed. No evidence of mitotic activity.	Spermatogonia the only germ cell observed. Tubules filled with cytoplasmic debris.	Tubules filled with cellular debris.
3 weeks	No spermatogonia present. Tubules small and filled with Sertoli cells.	No spermatogonia present. Tubules small and filled with Sertoli cells.	Most tubules depleted of spermatogonia. Some spermatids and immature sperm observed.	Tubules contained only Sertoli cells. Tubules atrophied.	Only Sertoli cells remained. Tubules atrophied.	Tubules contained only Sertoli cells.
5 weeks	Spermatogonia and primary spermatocytes present. Abnormally oriented immature sperm observed in some tubules.	Spermatogonia and primary spermatocytes present. Abnormally oriented immature sperm observed in some tubules	Spermatogonia and one or two rows of spermatocytes observed in most tubules.	Spermatogonia and spermatocytes noted in tubules. Mitotic figures numerous.	Evidences of recovery noted. Only spermatogonia and primary spermatocytes present in tubules.	Many tubules contained spermatogonia and primary spermatocytes. Some contained spermatids and abnormally oriented sperm. Some tubules contained only spermatogonia.

* Two treatments of 50 r each given 1 week apart.

Table 11. Effect of X-irradiation of the testes of twelve-week-old White Leghorn cockerels

Time after irradiation	X-Ray dosage in r units					
	50	50 + 50*	100	200	300	500
1 week	Secondary spermatocytes and spermatids numerous. Scattered sperm observed in some tubules, which were usually abnormally oriented.	Sloughing of cellular material to center of tubule noted. Sperm oriented in an abnormal manner.	Cell membranes difficult to distinguish. Central area of many tubules contained only Sertoli cell cytoplasm. Central area of others contained spermatids.	Many tubules were sloughing their cells. In other tubules cells were greatly disorganized.	Most tubules were filled with a disorganized mass of necrotic cells. Other tubules contained only Sertoli cells.	Seminiferous tubules in a highly variable condition. Some tubules had only Sertoli cells. Most tubules contained nearly all stages of spermatogenesis but were highly disorganized.
3 weeks	Many multinuclear giant cells observed. All germ cells had some cytoplasm around them causing a loose appearance of the tissue in the tubules.	Many secondary spermatocytes noted. Mitotic activity observed. Tubules showed some cellular reorganization with sperm uniformly oriented.	Most tubules had sloughed their necrotic cells. Many multinuclear giant cells observed.	Some tubules had many spermatogonia and primary spermatocytes. Other tubules had only a basal row of cells and had not started cellular regeneration.	Most tubules had a basal row of cells. The central areas of most tubules had various stages of spermatogenesis.	Tubules were still in a very variable condition. Some tubules contained only Sertoli cells. Most tubules contained all stages of spermatogenesis except sperm.
5 weeks	Cellular reorganization in process. Number of spermatogonia subnormal. Many secondary spermatocytes observed. Many organized groups of sperm noted.	Considerable mitotic activity noted. Few spermatids were observed but sperm, associated with Sertoli cells, were present in all tubules.	Many tubules contained small groups of sperm. Many tubules contained only spermatogonia and spermatocytes.	Some tubules contained all stages of spermatogenesis but only a few contained sperm. Other tubules contained only spermatogonia and primary and secondary spermatocytes.	Tubules were small in diameter. Contained only one row of spermatogonia and Sertoli cells.	All tubules recovering rapidly. Some tubules had small groups of sperm lining the lumina.

* Two treatments of 50 r each given 1 week apart.

Table 12. The effect of X-irradiation on the testes of 18-week-old White Leghorn cockerels

Time after irradiation	X-Ray dosage in r units					
	50	50 + 50*	100	200	300	500
1 week	Difficult to distinguish from controls. Some decrease in mitotic activity noted.	Most tubules were in a variable condition. In some tubules cellular membranes were only faintly visible. The amount of cellular sloughing was quite variable.	Most tubules were in a variable condition. Some tubules contained all stages of spermatogenesis but in an abnormal state. Other tubules contained only Sertoli cells and cellular debris.	Most tubules contained cellular material which resembled a molten mass that had shifted to occupy all the available space. Sperm were located in the central tubular area and were abnormally arranged.	All tubules were filled with a mass of cellular material. Cell membranes were distinct in tubules but not in others. In those tubules containing sperm, the sperm were highly disorganized.	All tubules filled with cellular material resembling a molten mass. Sperm present were in the disorganized mass in the center of the tubules.
3 weeks	Cells in tubules were slightly disorganized. Sperm were either scattered in the lumina of the tubules or were indiscriminately scattered in the walls of the tubules.	Most tubules were quite variable. In most tubules the cells had shifted inward until the lumina had become obliterated. Mitosis was evident but the sperm were usually abnormally oriented. A few tubules had sloughed their cellular material and only Sertoli cells remained.	Most tubules were still in a variable condition. Mitosis was evident in some tubules. Cellular necrosis and sloughing was also noted in many tubules.	Conditions observed were: (1) tubules small in diameter but sloughing completed; (2) tubules large in diameter but beginning sloughing; (3) tubules large in diameter and filled with spermatogonia and primary spermatocytes.	Tubules were found to be in three stages of recovery: (1) tubules small and all cells sloughed except basal row; (2) large tubules containing two or three rows of spermatogonia in mitosis; (3) large tubules containing advanced stages of spermatogenesis including sperm.	Some tubules had just finished sloughing their cellular material. Most tubules were in the process of recovery and were quite variable due to the difference in rate of recovery among the various tubules.
5 weeks	Some tubules appeared almost normal. Most tubules had scattered sperm or sperm concentrated in small groups in an unorganized manner.	Many tubules were large and filled with germ cells in mitosis. Other tubules were small and contained only Sertoli cells.	Most tubules were in a variable condition. Cellular mitosis and sperm maturation were noted along with cellular atrophy and sloughing.	All tubules were in the process of recovery. The stage of recovery depending upon the length of time recovery had been in progress.	All tubules producing germ cells, but most had not progressed beyond the secondary spermatocyte stage. Small groups of sperm observed in some tubules.	All tubules in the process of recovery but cellular disorganization quite evident. Many tubules were in a syncytial condition with recovery rather slow in comparison to other irradiation levels.

* Two treatments of 50 r each given 1 week apart.

reduction was followed by a very slow recovery rate, particularly in the older birds.

(6) The total leucocyte count resembled that noted for the lymphocytes. There was an immediate drop in the total leucocyte count, followed by a gradual recovery toward the normal level.

(7) There was an indication of a slight stimulatory effect on the erythrocyte count and hemoglobin level at the higher irradiation dosages (200–500 r), followed by a rather rapid return to the normal level.

(8) There was no cellular damage to the thyroid gland which could be attributed to irradiation.

(9) There was no cellular damage observed in the pancreas which could be attributed to X-irradiation.

(10) No cellular damage was observed in the anterior pituitary which could be attributed directly to irradiation. However, β- (basophil) cells containing small vacuoles characteristic of castrate cells were noted in the pituitaries of cockerels irradiated at twelve and eighteen weeks of age with 200, 300 and 500 r.

(11) The exposure of cockerels to whole body X-irradiation caused an increase in the numbers of erythrocytes in the red pulp of the spleen, regardless of the age of the bird or the dosage level used.

Heterophils were noted in the red pulp during the third and fifth weeks after treatment, and seemed to be related to age and dosage level with larger numbers being found in the older birds given higher doses of X-rays.

The lymphocytes were very sensitive to irradiation and were least in evidence in the spleen during the first week after irradiation. The process of recovery from the lymphocytopenia was evident during the third week after treatment but recovery rate appeared to be related to age and dosage level with recovery being fastest in the younger birds. At the higher dosage levels (300–500 r) reticular elements were the only cellular elements remaining the first week after irradiation.

(12) The effects of whole-body X-irradiation on the testes of young cockerels, as judged by histological appearance, was most evident the third week after treatment. Irradiation damage was manifested in the seminiferous tubules as follows: (1) sperm scattered among the cells composing the walls of the tubules, (2) a decrease in the number of sperm attached to the Sertoli cells, (3) sperm oriented in an abnormal manner, (4) the presence of cell necrosis, and (5) a massive sloughing of cells in the tubules.

The amount of observable damage which occurred depended upon: (1) the quantity of irradiation, (2) the age of the bird at the time of irradiation, and (3) the stage of spermatogenesis present in the seminiferous tubules at the time of irradiation.

Spermatogonia were the most sensitive and sperm the most resistant germ cells to irradiation. Sertoli cells and the interstitial cells of Leydig were also very resistant to irradiation. The spermatogonial cells were severely damaged or killed but secondary spermatocytes and later stages continued their development to form sperm unless the dosage was of sufficient strength to cause massive sloughing or necrosis. Because the primitive germ cells were sensitive to irradiation there was an interval of time in which the seminiferous tubules were, in many cases, completely depleted of germ cells.

Multinuclear giant cells appeared in the lumina of the tubules of birds irradiated at 12 weeks of age. These cells appeared to be caused by a fusion of spermatids which then became associated with the Sertoli cell. The Sertoli cell cytoplasm with its associated and fused spermatids then separated from the remainder of the Sertoli cell and was observed in the lumina of the seminiferous tubules.

Acknowledgment—This research was supported in part by a grant, AT(40-1)-1307, from the Atomic Energy Commission for which grateful acknowledgment is extended.

REFERENCES

1. G. Bignomi, *Haematologica Arch.* **20,** 561 (1939).
2. R. M. Mulligan, *Proc. Soc. Exp. Biol., N.Y.* **48,** 607 (1941).
3. A. Nettleship, *Radiology* **42,** 64 (1944).
4. P. S. Henshaw, *J. Nat. Cancer Inst.* **4,** 477 (1944).
5. P. S. Henshaw, *J. Nat. Cancer Inst.* **4,** 485 (1944).
6. P. S. Henshaw, *J. Nat. Cancer Inst.* **4,** 513 (1944).
7. T. F. Dougherty and A. White, *Endocrinology* **39,** 370 (1946).
8. L. O. Jacobson, E. K. Marks and E. Lorenz, *Radiology* **52,** 371 (1949).

9. T. J. Haley and H. Harris, *Science* **111,** 88 (1950).
10. E. Eldred and B. Eldred, *Blood* **8,** 262 (1953).
11. W. E. Cornatzer, O. Engelstad and J. P. Davison, *Amer. J. Physiol.* **175,** 153 (1953).
12. A. M. Lucas and E. M. Denington, *Poultry Sci.* **36,** 1290 (1957).
13. T. S. P. Strangeways and F. L. Hopwood, *Proc. Roy. Soc.*, B **100,** 283 (1926).
14. M. Rosenbaum, *Wien. klin. Wschr.* **40,** 1315 (1927).
15. A. B. Dawson, *J. Exp. Zool.* **46,** 467 (1927).
16. S. L. Warren, *Physiol. Rev.* **8,** 92 (1928).
17. R. M. Oslund and A. Bachem, *Proc. Soc. Exp. Biol., N.Y.* **23,** 761 (1926).
18. H. Wintz, *Mschr. Geburtsch. u. Gynakol.* **78,** 428 (1928).
19. J. B. Gatenby and S. Wigoder, *Proc. Roy. Soc.* B **104,** 351 (1929).
20. R. Ferroux, Cl. Regaud and N. Somssonow, *C. R. Soc. Biol., Paris* **128,** 173 (1938).
21. C. K. Hu and C. N. Frazier, *Proc. Soc. Exp. Biol., N.Y.* **48,** 44 (1941).
22. A. B. Eschenbrenner and E. Miller, *J. Nat. Cancer Inst.* **6,** 343 (1946).
23. L. C. Fogg and R. F. Cowing, *Cancer Res.* **11,** 23 (1950).
24. H. I. Kohn, *Rad. Res.* **3,** 153 (1952).
25. L. C. Fogg and R. F. Cowing, *J. Exp. Cell Res.* **3,** 19 (1952).
26. L. C. Fogg and R. F. Cowing, *J. Exp. Cell Res.* **3,** 245 (1952).
27. S. L. Shaver, *Amer. J. Anat.* **92,** 391 (1953).
28. S. L. Shaver, *Amer. J. Anat.* **92,** 433 (1953).
29. P. B. Hawk, B. L. Oser and W. H. Summerson, *Practical Physiological Chemistry.* Blackiston, Philadelphia (1947).
30. M. P. Natt and C. A. Herrick, *Poultry Sci.* **31,** 735 (1952).
31. W. Bloom and M. A. Bloom, *Rad. Biol.* McGraw-Hill, New York (1954).
32. R. G. Murray, *Histopathology of Irradiation from External and Internal Sources.* McGraw-Hill, New York (1948).

Health Physics Pergamon Press 1961. Vol. 6, pp. 177–182. Printed in Northern Ireland

EFFECT OF CHELATING AGENTS ON THE REMOVAL OF YTTRIUM AND LANTHANUM FROM MAN

BETTY ROSOFF, SEYMOUR RITTER, KATHLEEN SULLIVAN, HIRAM HART and HERTA SPENCER-LASZLO

Division of Neoplastic Diseases, Montefiore Hospital, New York, N.Y.

(*Received* 9 *January* 1961)

Abstract—Yttrium chelate excretion studies have been conducted in this laboratory stimulated by the interest in the removal of fission products from man. Since diethylenetriaminepenta-acetic acid (DTPA) has a high affinity for yttrium and is almost quantitatively excreted in 24 hr, the effectiveness of DTPA in removing yttrium from the body was studied in eight patients. Yttrium was injected intravenously as the nitrilotriacetate (Y^{90}-NTA), a weak chelate, and 24 hr later, 500 mg DTPA was infused daily on four successive days. The Y^{90} excretion was then compared with the results obtained with equivalent amounts of EDTA. The effectiveness of 2 g of either chelate was also studied. The infusions of 500 mg DTPA increased the yttrium excretion from four to eight times over the base line excretion. DTPA was more effective than EDTA and a fourfold increase of either chelate did not result in an appreciably higher excretion of the metal chelate than was achieved with the 500 mg dose. In studies carried out in two patients, doses of 100 mg DTPA were found to be less effective than 500 mg of this chelating agent. Similar studies of the removal of La^{140} indicate that DTPA is also more effective than EDTA in removing this rare earth isotope from man. The significance of these decontamination studies and their possible application for fission product removal will be discussed.

INTRODUCTION

In view of the increasing use of nuclear energy in industry and medicine, contamination of the human body by radioisotopes may be one of the newer occupational hazards. The development of methods of decontamination, have, therefore, received increasing attention in recent years. Studies on the modification of tissue distribution and excretion of radioactive rare earth fission products administered in ionic and chelated form have been carried out in this laboratory.[1–3] A very wide range of excretion for different chelates was noted extending from minimal to almost quantitative excretions. The observation was made that the magnitude of excretion of the radioactive rare earth correlates with the stability constant of the rare earth chelate.[4] *In vitro* studies have shown a correlation between the stability constant of a metal chelate and its filterability: the ultrafiltration of strong metal chelates from serum indicated preferential binding of the metal to the chelate as opposed to binding of the metal by body competitors.[5–7] In the search for agents which may be effective in removing yttrium from the body, chelating agents of the polyamino acid type have been selected for study since they have a strong affinity for yttrium and since the urinary excretion of these chelating agents was found to be high. When diethylenetriaminepenta-acetic acid (DTPA) was injected intravenously as the yttrium chelate, labeled with Y^{90}, the urinary excretion of Y^{90} was found to be almost 100 per cent in 24 hr.[4] The effectiveness of this compound in removing yttrium was, therefore, investigated and compared with the more conventionally used chelating agent ethylenediaminetetraacetic acid (EDTA). Some of these data were recently presented[8] and the details of these studies form the basis of this communication.

MATERIALS AND METHODS

Table 1 lists the patients and the type of studies performed. Thirty-five studies were carried out in

Table 1. *List of patients studied*

Patient	Age,	Sex	Diagnosis	Rare earth isotope	Chelating agent (mg)*	
					DTPA	EDTA
1	75	M	Osteoarthritis	Yttrium-90	117 586 2340	100 500 2000
2	65	M	Paget's disease	Yttrium-90	117 586 2340	100 500 2000
3	60	F	Carcinoma of rectum	Yttrium-90	586 2340	500 2000
4	62	F	Metastatic bone disease	Yttrium-90	586	500 2000
5	60	F	Multiple myeloma	Yttrium-90	586	500 2000
6	60	F	Carcinoma of lung	Yttrium-90	586	500
7	51	M	Carcinoma of lung	Yttrium-90	586	500
8	73	F	Multiple myeloma	Lanthanum-140	586† 2340†	500† 2000†
9	52	M	Hypoparathyroidism	Lanthanum-140	586 2340	500 2000

This study was carried out under Contract AT (30–1) 1763 with the United States Atomic Energy Commission.

* Each amount of the chelate was infused on four successive days unless otherwise indicated.

† This amount given on two successive days only.

nine patients. All patients had normal renal function as determined by clinical laboratory tests. The removal of yttrium was studied in seven patients and that of lanthanum in two patients. In order to prevent the formation of yttrium hydroxy colloids, yttrium was injected intravenously in the form of the weak chelate, yttrium-90 nitrilotriacetate (Y^{90}-NTA). Patients in whom studies of yttrium decontamination were carried out received five tracer doses of Y^{90}-NTA (10 μc Y^{90}). The urinary excretions of Y^{90} of the first 5 days following the administration of the first tracer dose served as the control study. Decontamination studies were then started 24 hr following the intravenous injection of the second tracer dose. Five hundred milligrams of Na-EDTA were infused over a 4-hr period on each of the four successive days and the urinary excretion of Y^{90} was again determined for a total of 5 days. Similarly, 586 mg of Na-DTPA (molar equivalent amount of the infused Na-EDTA) were infused on four successive days starting 24 hr following the injection of the third tracer dose of Y^{90}-NTA. In order to study the effectiveness of different amounts of EDTA and of DTPA on the removal of yttrium, 2 g of each of these chelates were infused on four successive days starting 24 hr following the intravenous injection of the fourth and fifth tracer, respectively. The effectiveness of much smaller amounts of either chelate, 100 mg Ca-DTPA or Ca-EDTA, respectively, was also tested in two patients in a similar manner (patients 1 and 2). The removal of La^{140} by DTPA and EDTA was studied in patients 8 and 9. The baseline excretions of tracer

doses of La^{140} given as the chloride were determined in 5 days. Infusions of 500 mg Ca-EDTA, and Ca-DTPA, respectively, were given for 2 days each starting 24 hr following the injection of $La^{140}Cl_3$.

Solutions

Lanthanum and yttrium solutions were prepared by dissolving the respective rare earth oxide in dilute HCl and labeling the resultant YCl_3 with Y^{90}* and $LaCl_3$ with La^{140}.† Y^{90}-NTA was prepared by combining $Y^{90}Cl_3$ with nitrilotriacetic acid in a molar ratio of 1:1.2 of the metal to the chelate. The tracer doses of the radioactive rare earths contained 1 mg stable yttrium or 1.5 mg stable lanthanum, respectively. The chelate solutions were prepared by dissolving the appropriate amounts of EDTA and DTPA in dilute NaOH to form the sodium salt. Calcium disodium salts were prepared by adding calcium chloride to the chelating agent in a 1:1 molar ratio. All solutions were made up in normal saline and adjusted with dilute NaOH to pH 7.5. The infusion solutions were prepared by adding the required amounts of the respective chelate to 250 ml of 5 per cent glucose in water.

Counting methods

In each study, a standard equal to the dose injected into the patient was prepared by drawing up the same volume of the injection solution using the same syringe and needle. This volume was then delivered into a volumetric flask and after proper dilution, aliquots of this solution were assayed for radioactivity. The Y^{90} standard and the Y^{90} excretion in urine were radioassayed by delivering 1-ml or 2-ml aliquots of the respective fluid onto metal planchets, dried under an infrared lamp and then counted using a standard end window Geiger tube. The La^{140} content of both the standard solution and of the urine was counted in a well-type scintillation counter, using 1- or 5-ml aliquots.

* Y^{90} was prepared by neutron activation of Y_2O_3 at Brookhaven National Laboratory.

† La^{140}, a fission product, was obtained from Oak Ridge National Laboratory.

RESULTS

Fig. 1 shows an example of the comparative Y^{90} excretion following four successive infusions each of 500 mg and 2000 mg of EDTA and of four infusions of equimolar amounts of DTPA. Both chelates resulted in marked enhancement of Y^{90}, varying from four to eight times the control excretion. However, DTPA is more effective than EDTA in removing yttrium when either amount of the chelate is used. For instance, the cumulative excretion of Y^{90} is twice as high following the four infusions of 586 mg DTPA than of an equimolar amount of EDTA. Four successive infusions of 586 mg DTPA are as effective as four infusions of 2340 mg DTPA. EDTA was somewhat more effective in the 2000 mg level than at the 500 mg level.

Table 2 shows the 4-day cumulative excretion of Y^{90} expressed in per cent of the administered dose remaining in the body 24 hr following the administration of Y^{90}-NTA. The average control excretion of Y^{90}-NTA is 3.8 per cent in 4 days. When Na-EDTA (500 mg and 2000 mg)

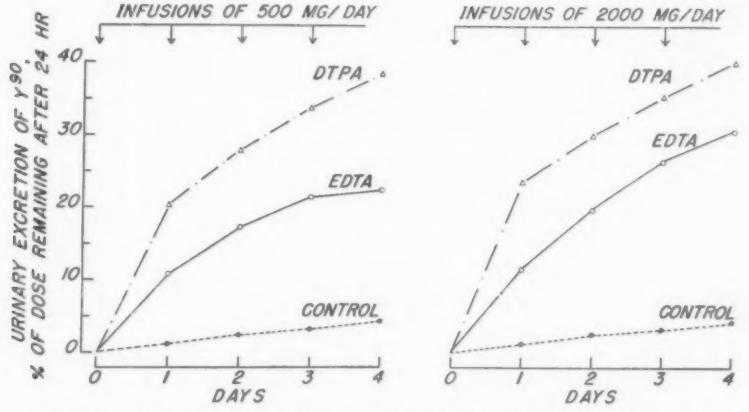


FIG. 1. Yttrium removal from man by Na-EDTA and Na-DTPA infusions. (Y^{90}-NTA was injected 24 hr prior to the infusions.)

Table 2. Removal of yttrium (Y^{90}) by Na-EDTA and Na-DTPA in man

Patient	4-day cumulative urinary excretion of Y^{90} in % dose remaining 24 hr following Y^{90}-NTA injection				
	Control (Y^{90}-NTA)	Na-EDTA		Na-DTPA	
		500 mg	2000 mg	586 mg	2340 mg
1	5.0	35.8	42.2	45.1	43.5*
2	0.54	23.5	22.1	38.3	32.9*
3	4.4	22.3	30.6	38.3	40.1
4	5.5	28.0	24.2†	41.5	—
5	4.9	31.4	36.7	31.5	—
6	1.5	20.2†	—	27.9	—
7	5.1	32.3	—	38.9	—
Average 3.8		27.6	31.2	37.3	38.8

* Patients 1 and 2 received Ca-DTPA.
† Minimal values because of undetermined urine losses.

was given, 27.6 per cent and 31.2 per cent, respectively, were excreted in the same period of time. Na-DTPA resulted in a tenfold increase of the control excretion: 37.3 per cent and 38.8 per cent of the dose remaining was excreted. The data shows that a fourfold increase of either chelating agent did not result in any appreciably higher excretion of yttrium. When equimolar amounts of each chelate was used, Na-DTPA proved more efficient than Na-EDTA.

Fig. 2 shows the comparative Y^{90} excretion of the two patients following four infusions containing 100 mg, 500 mg and 2000 mg DTPA, respectively. In each of the two patients when 100 mg DTPA was infused, the Y^{90} excretion increased approximately tenfold as compared to

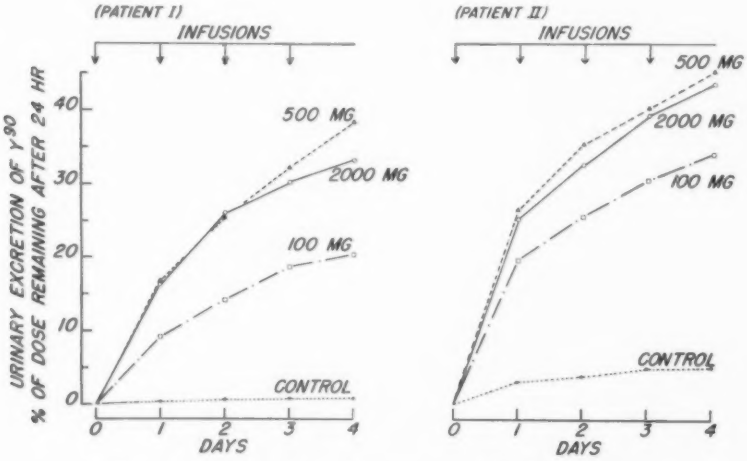


FIG. 2. Yttrium removal from man by Ca-DTPA infusions. (Y^{90}-NTA was injected 24 hr prior to the infusions.)

the control excretion. However, infusions of 100 mg DTPA were not as effective as 500 mg DTPA. This figure shows again that the infusions of 2000 mg DTPA were not more effective than the 500 mg dose.

Fig. 3 shows the La^{140} excretions of two patients following two infusions each of 500 mg EDTA and of equimolar amounts of DTPA. The La^{140} excretion increased approximately three times when EDTA was infused. The administration of DTPA resulted in a nine-to-tenfold enhancement of the La^{140} excretion.

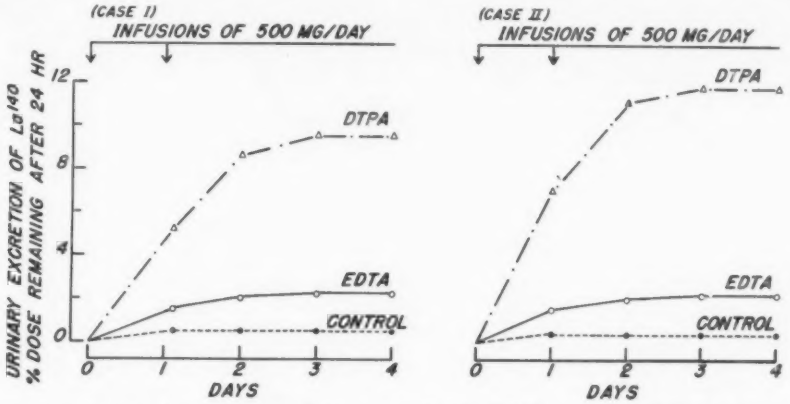


FIG. 3. Lanthanum removal from man by Ca-EDTA and Ca-DTPA infusions. ($La^{140}Cl_3$ was injected 24 hr prior to the infusions.)

DISCUSSION

The results indicate that chelating agents are effective in removing rare earth isotopes from the body. If a chelate is administered 24 hr after the injection of the rare earth isotope, up to 40 per cent of yttrium and up to 12 per cent of lanthanum remaining in the body can be removed. Presumably the major part of the isotope is present in the circulating blood, the extracellular fluid and soft tissue at this time. SNYDER *et al.*[9] have demonstrated that rare earths localize in the liver and in fact cause fatty infiltration. Similar observations were made in this laboratory several years ago.

GRAUL and HUNDESHAGEN[10] recently reported studies of yttrium distribution in guinea-pigs, and state that yttrium-90 administered as the chloride is not deposited in bone in the first 8 days after the administration of the isotope. The rare earth isotope may, therefore, be available for removal in this period of time. Studies on the mechanism of the uptake of yttrium by bone have been reported.[11] The studies reported here have shown that effective decontamination can be accomplished in this phase. Whether yttrium can be removed after it is deposited in bone will have to be determined in subsequent experiments. Tissue distribution studies carried out with Y^{90}-NTA in mice reveal that most of the yttrium is concentrated in liver and spleen 24 hr after the injection of the radioisotope.[12] DTPA was shown to be effective in studies of the removal of radio-yttrium from mice and the administration of this chelate again resulted in greater removal of Y^{90} than EDTA.

FRIED *et al.*[13] have shown that the excretion of plutonium in rats can be enhanced by the chelating agents EDTA, DTPA and BAETA, bis (2-aminoethyl) ether tetra-acetic acid. These investigators were able to promote an increase of plutonium excretion even 22 days after its administration which indicates that it is possible to remove this metal from bone due to the high stability constant of this chelating agent with plutonium.

Rare earths have been known to combine with nucleic acids, and therefore removal of the isotope might involve breaking the metal nuclear protein bonds. *In vitro* studies carried out in this laboratory indicate that the yttrium chelate bond

is stronger than the binding of yttrium to serum proteins.[6–7] Studies on the competitive binding of yttrium chelates and DNA are in progress.

The work presented here has shown that diethylenetriaminepenta-acetic acid (DTPA) is an effective decontaminating agent for yttrium and lanthanum in man. EDTA and DTPA have also been used to remove lead[14] and cerium[15] from the mammalian organism. It is likely that these chelating agents may become of practical value in the decontamination of other heavy metals and their radionuclides. Ca-DTPA would be the agent of choice since Na-DTPA might interfere with calcium homeostasis. The toxicity of EDTA was reported to be minimal in man[16–17] and in subsequent studies DTPA was given in doses up to 4 g per day and was well tolerated. The usefulness of chelating agents of the polyamino acid type is apparent in accidental exposure to radioisotopes. Yttrium-90 is finding a more widespread use in the treatment of pleural and ascitic effusions due to cancer[18] and, in cases of accidental entry of this radioisotope into the blood stream, DTPA may be of value for its removal.

SUMMARY

(1) The chelating agents EDTA and DTPA have been used for the removal of yttrium and lanthanum from man.

(2) Relatively small doses of either chelating agent enhance considerably the urinary excretion of yttrium and lanthanum.

(3) Ca-DTPA was found to be a more effective decontaminating agent than EDTA.

(4) A study of different dose levels of DTPA showed that a plateau of yttrium removal was reached at the 500 mg level.

(5) Ca-DTPA may have practical application for the removal of fission products in man, especially in cases of acute exposure.

REFERENCES

1. R. Lewin, H. E. Hart, J. Greenberg, H. Spencer, K. G. Stern and D. Laszlo, *J. Nat. Cancer Inst.* **15,** 13i (1954).
2. H. E. Hart, J. Greenberg, R. Lewin, H. Spencer, K. G. Stern and D. Laszlo, *J. Lab. Clin. Med.* **46,** 182 (1955).
3. R. Lewin, H. Spencer, K. G. Stern and D. Laszlo, *Proc. Amer. Ass. Cancer Res.* **1,** 33 (1953).
4. H. Kroll, S. Korman, E. Siegel, H. E. Hart, B. Rosoff, H. Spencer and D. Laszlo, *Nature, Lond.* **180,** 919 (1957).
5. R. Lewin, B. Rosoff, H. E. Hart, K. G. Stern and D. Laszlo, In *Advances in Radiobiology* p. 298. Oliver and Boyd, Edinburgh (1957).
6. B. Rosoff, R. Lewin, H. E. Hart, G. L. Williams and D. Laszlo, *Arch. Biochem. Biophys.* **78,** 1 (1958).
7. B. Rosoff, *Ann. N.Y. Acad. Sci.* **88,** 479 (1960).
8. B. Rosoff, S. Ritter, K. Sullivan and H. Spencer, *Fed. Proc.* **18,** 327 (1959).
9. F. Snyder, E. A. Cress, N. Stephens and G. C. Kyker, *Fed. Proc.* **18,** 327 (1959).
10. E. H. Graul and H. Hundeshagen, *Int. J. Appl. Radiat. & Isotopes* **5,** 243 (1959).
11. G. R. Martin, B. J. Mulryan and W. F. Neuman, Abstract presented at Pharmacology Society Meetings, August (1958).
12. E. Siegel and H. Spencer–Laszlo, Unpublished data.
13. J. F. Fried, E. H. Graul, J. Schubert and W. M. Westfall, *Atompraxis* **5,** 1 (1959).
14. F. Rieders, In *Metal-Binding in Medicine* p. 143. Lippincott, Philadelphia (1960).
15. A. Catsch and D. K. Le, *Nature, Lond.* **180,** 609 (1957).
16. H. Spencer, J. Greenberg, E. Berger, M. Perrone and D. Laszlo, *J. Lab. Clin. Med.* **47,** 29 (1956).
17. M. J. Seven, In *Metal-Binding in Medicine* p. 95 Lippincott, Philadelphia (1960).
18. E. Siegel, H. E. Hart, M. Brothers, H. Spencer and D. Laszlo, *J. Amer. Med. Ass.* **161,** 499 (1956).

Health Physics Pergamon Press 1961. Vol. 6, pp. 183–188. Printed in Northern Ireland

THE BILIARY EXCRETION OF RADIUM*

JOHN B. HURSH and ARVIN LOVAAS
Department of Radiation Biology, University of Rochester, Rochester, New York

(*Received* 11 *May* 1960)

Abstract—Two female dogs were prepared with in-dwelling gall bladder catheters and after recovery from the operation received an intravenous injection of 1 μc of radium. Twenty-four-hour samples of bile, urine and feces were collected for total periods of from 16 to 46 days. It was found that the daily biliary outflow of radium contributes only a few per cent to the total radium excreted per day. The relative role of other modes of contribution to the fecal output is discussed.

INTRODUCTION

FECAL excretion is an important means for removal of radium from the organism. Thus, in dogs, as seen in Fig. 1, approximately 70 per cent of the total excretion is by the feces, independent of time after injection and therefore of the blood radium level. Although the biliary discharge into the gut has been implicated as a major source of the radium found in the feces, the existing data do not allow a quantitative estimate of the importance of its role. Acute sacrifice experiments performed by SALANT and MEYER[2] showed that the bile collected from radium injected dogs was "radioactive". AUB *et al.*[3] measured radium in the gall stones found upon autopsy of a radium patient.

In order to evaluate the daily biliary secretion of radium into the gut, two dogs were prepared with in-dwelling gall bladder catheters, received an intravenous injection of radium, and 24-hr collection of bile, urine and feces were made.

PROCEDURE

Two adult female mongrel dogs were prepared according to the closed sterile fistula method of ROUS and MCMASTER[1], with the difference that, instead of cannulating the common bile duct, a mushroom catheter was inserted into the gall bladder. The tube leading from the catheter was brought to the outside through a stab wound just posterior to the rib cage. The tube is joined to one arm of a glass T, with the other arms connected to a 350-ml rubber storage balloon and a sterile collection vent, respectively. The collection apparatus is protected by a fiber cup-shaped shield supported in place by a canvas harness.

After the operation the dogs were observed until the daily volume of bile output was judged to be in the normal range. At this time 1 ml of a solution containing 1.0 μc of radium chloride at the pH 6.0 was injected into the jugular vein. An equal volume of the injection solution was discharged into a bottle containing 100 ml of 5 per cent HCl acid solution and designated the dummy injection. The dogs were placed in metabolism cages and daily collections were made of urine, feces and bile as indicated in

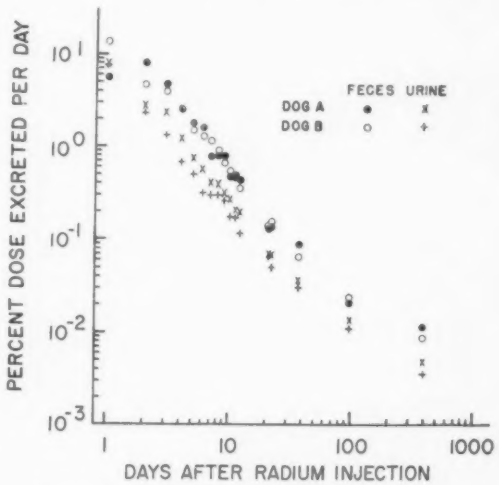


FIG. 1. The fecal and urinary output[1] of two dogs after an intravenous injection of radium at zero time.

* This report is based on the work performed under contract with the U.S. Atomic Energy Commission at the University of Rochester, Atomic Energy Project, Rochester, New York.

Table 1. Both dogs continued well throughout the experiment. There was no indication of infection and no change in color or odor of the bile collected. At certain intervals (see Table 1) bile salt pills were administered by mouth. Dog *C* was sacrificed 56 days after the bile fistula operation. Dog *D* received a second radium injection 27 days after the first injection and was sacrificed 88 days after the operation.

The radium content of the 24-hr samples of bile, urine and feces was measured either by the radon emanation method (dog *C*) or by a γ-counting, sodium iodide crystal technique (dog *D*).

The urine and bile samples from dog *C* were wet ashed in concentrated nitric acid to a white soluble salt, taken up in 200 ml of 0.5 N HCl acid, and de-emanated in the equipment illustrated in Fig. 2. The tower was sealed and allowed to collect radon for a known period of time. The accumulated radon was swept out with tank nitrogen and transferred to an ionization chamber. The ionization current was measured in a vibrating reed electrometer and radium content of the original sample was calculated in the usual way. A sub-dilution of the dummy injection was similarly analyzed omitting the digestion step.

The fecal samples were dry ashed at 600°C, further digested with concentrated nitric acid, and the salt was taken up in 200 ml of 0.5 N hydrochloric acid. If an insoluble residue remained, it was filtered out and saved. The filtrate was then processed and the ionization current measured as described above. The residue and filter paper were transferred to a 13-mm by 100-mm ignition tube and ashed. Close-fitting glass plungers were inserted into the tubes so that approximately 1 ml of space containing the ash remained at the bottom of the tube. The plunger was sealed in place with wax, and the tube set aside for 3 weeks to allow radon and its daughters to grow to equilibrium. The γ-sample activity was then counted in a sodium iodide crystal well. The result was converted to its radium equivalent on the basis of the counting rate of a radium standard similarly

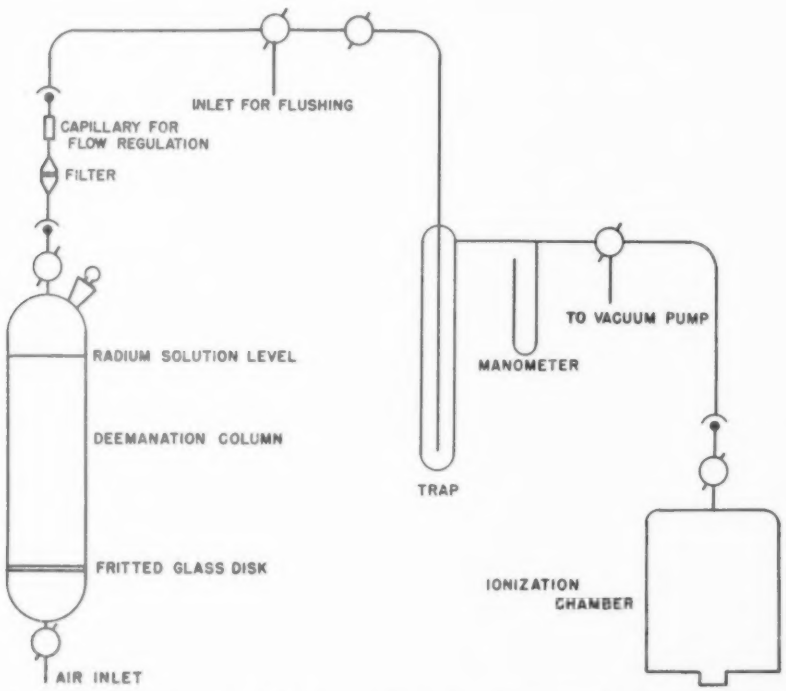


FIG. 2. Schematic presentation of the apparatus for de-emanation of radium solutions. The de-emanation column has a total volume of 250 ml. The fritted disks are of medium porosity. The capillary used is about 7 mm long and has an internal diameter of 0.18 mm. The trap removes water vapor and is cooled by an acetone–solid CO_2 mixture.

contained. The results obtained on the two fractions were combined to yield the total radium content of the fecal sample.

The relative convenience, sensitivity and reproducibility of the scintillation crystal measurement technique used for the fecal insoluble residues brought about the decision to use this method exclusively for the bile, urine and fecal samples collected during the two experimental periods on dog *D*. To this end fecal samples were dried on a hot plate, ashed in a muffle furnace, and the entire sample transferred to one or more glass vials. The vials were chosen to fit loosely in the crystal well and sealed in such a way that the sample space was about 1 ml volume. The urine and bile samples were wet ashed to an extent necessary to reduce the volume to 1 ml and transferred to similar vials, which were sealed. The vials were set aside for sufficient time for the radon and its daughters to grow to near equilibrium and counted in the well counter. A series of six 1-ml aliquots of sub-dilutions of each of the two dummy injections were similarly sealed into vials and counted. Counts during the growth period served to identify any vials with leaks.

RESULTS

The results presented in Table 1 indicate that the radium outflow through the bile constitutes only a few per cent of the daily total excretion. The fecal excretion of radium for days 1 and 2, dog *C*, is exceptionally low (see Fig. 1 and corresponding days for dog *D*). Although no reason can be assigned for this, it is possible that the interval of 1 week, while sufficient to allow full recovery of the bile flow mechanism did not permit full recovery of the other channels by which radium enters the gut. On the other hand, dog *C*, with an 11-day recovery period shows the usual amount of radium excreted in both feces and urine. In general, the individual values show irregular variations from day to day, and there are infrequent instances when no sample of urine or feces were voided. Whether the absence of bile in the gut caused this irregularity in fecal output, can only be conjectured. It has been shown that obstruction of the biliary ducts (Canonico and Mann[5]) is accompanied by a diminished intestinal motility.

At the times indicated in Table 1, bile salt pills (0.325 g dried purified bile per day) were given by mouth. This dose is to be considered supplementary and is not sufficient to substitute for the daily loss of bile salt in the fistula dog. Smith *et al.*[6] found that on their standard diet a fistula dog put out about 100 mg of bile salt per kg body weight. Since our dogs weighed 10 kg, it might be inferred that their output was about 1 g of bile salt per day. It will be seen that with the possible exception of days 44–46, dog *C*, the bile salt pills did not appear to bring about any increase in the bile volume collected. Despite this demand upon the dogs' mechanism to produce bile salt, it may be noted that the average daily bile volume and the radium excretion by this pathway, show no decrease for the second experimental period, dog *D*, as compared with the first experimental period.

DISCUSSION

Although the fecal radium output is 70 per cent of the total excreted, the experimental results described above show that the radium flow in the bile accounts for only a relatively small fraction. Therefore it may be concluded that the major delivery route of radium into the gut is either the salivary out-flow, the gastric or the intestinal cell secretion, or a combination of these pathways.

Little information is available on radium secretion by the salivary glands. A single incidental observation made in this laboratory showed that saliva collected as a mouth drip from a dog 5 hr after an intravenous injection of radium, yielded a radium concentration equal to 56 per cent of the radium concentration of a plasma sample drawn at the same time. It is of some interest that the ultrafilterable fraction of dog plasma radium has been found to average about 69 per cent.[5] Measurement[6] of radium's chemical analog, calcium, shows a ratio of normal salivary calcium to serum calcium of 60 per cent in man. These observations suggest that, in respect to radium and calcium, the saliva may be a plasma ultrafiltrate with no resorption of these ions. If it is assumed that the 24-hr salivary output of a 10-kg dog would be no more than about 500 ml (i.e. approximately half that of man) a daily salivary "clearance" of radium into the gut could be set at no more than about 300 ml of plasma per day. This clearance is small with respect, for instance, to the radium clearance by the dog kidney, variously measured[5] at from 1500 to 3000 ml per day. Pending more

Table 1. *Biliary elimination of radium*

Time (days)	Bile volume (ml)	Bile (% dose/24 hr)	Feces (% dose/24 hr)	Urine (% dose/24 hr)	Total % dose	Bile % total
			Dog *C*			
1	50	0.56	0.07	6.6	7.23	7.7
2	105	0.29	0.22	2.6	3.11	9.3
3	95	0.16	—	NS	—	—
4	67	0.12	5.62	2.01	7.75	1.55
5	115	0.098	1.89	0.81	2.80	3.5
6	68	0.061	0.63	0.61	1.30	4.7
7	52	0.046	0.65	0.21	0.91	5.1
8	118	0.047	0.24	0.44	0.73	6.4
9	98	0.040	0.93	0.31	1.28	3.1
10	38	0.020	0.12	0.26	0.40	5.0
11	150	—	0.32	0.24	0.56	—
12	98	0.051	0.64	0.23	0.92	5.5
13	93	0.022	NS	0.15	(0.17)	(12.9)
14	104	0.022	0.18	0.16	0.36	6.1
21	110	0.011	0.32	0.093	0.42	2.6
37	20	0.002	0.08	0.031	0.113	1.8
38	99	0.006	0.12	0.049	0.175	3.4
39	59	0.005	—	0.0055	(0.06)	(8.3)
44*	112	0.006	0.21	0.037	0.253	2.4
45*	113	0.006	0.22	—	(0.226)	2.7
46*	115	0.005	0.13	0.064	0.199	2.5
			Dog *D*(1)			
1	104	0.34	9.54	7.12	17.0	1.7
2	87	0.21	11.00	4.43	15.64	1.3
3	68	0.015	3.88	3.12	7.01	0.5
4	—	0.06	4.07	0.91	5.04	1.2
5	44	0.02	NS	0.57	(0.59)	(3.4)
6	43	0.02	NS	0.52	(0.54)	(3.7)
7*	55	0.05	2.06	0.05	2.16	2.3
8*	67	0.06	1.61	1.19	2.86	2.1
9*	65	0.03	1.93	0.59	2.55	1.2
10*	51	0.02	1.34	0.41	1.77	1.1
11*	34	0.018	0.24	0.26	0.52	3.5
12*	37	0.020	1.51	0.44	1.97	1.0
13*	20	0.013	NS	—	(0.013)	—
14	37	0.0082	0.76	0.53	1.30	0.6
15*	21	0.02	NS	0.18	(0.20)	(10)
			Dog *D*(2)			
1	68	0.158	9.29	7.83	17.28	0.9
2	68	0.129	11.5	5.16	16.79	0.77
3	72	0.10	5.96	1.67	7.73	1.3
4	45	0.052	1.38	1.09	2.52	2.0
5	—	0.135	3.91	1.28	5.32	2.5
6	48	0.048	2.24	0.97	3.26	1.5

Table 1—(*contd.*)

Dog *D*(2)

Time (days)	Bile volume (ml)	Bile (% dose/24 hr)	Feces (% dose/24 hr)	Urine (% dose/24 hr)	Total % dose	Bile % total
7*	68	0.063	0.76	0.46	1.28	4.9
8	77	0.050	2.48	0.44	2.97	1.7
9*	78	0.043	1.08	1.16	2.28	1.9
10	40	0.012	1.05	NS	(1.06)	(1.1)
11*	78	0.035	1.14	0.72	1.89	1.9
12	71	0.032	1.45	0.55	2.03	1.6
13*	43	0.022	0.73	—	(0.75)	(2.9)
14	47	0.015	NS	0.12	(0.13)	(11.5)
15*	42	0.040	NS	0.27	(0.31)	(12.9)
16	71	0.018	2.20	0.25	2.47	0.73

* 0.325 g of dried purified ox bile administered orally on the days marked.

satisfactory data on salivary radium content, the salivary outflow may be tentatively regarded, together with the biliary outflow, as a minor source of fecal radium.

The relative importance of other means by which radium may enter the gut may, to some degree, be estimated from the early experiments by SALANT and MEYER[2]. These authors performed acute experiments on dogs, injecting radium subcutaneously and isolating various sections of the gut by ligatures. After an appropriate interval the dogs were sacrificed and the radium content of the gut sections and their contents were estimated by the relatively crude measurement techniques available at that time. They found the stomach and its contents as well as the large intestine and its contents "inactive". The small intestine was inactive, but its contents were "active". They concluded that the kidney, liver and small intestine eliminate radium.

The same general method has recently been used by SINGER *et al.*[9] to investigate the endogenous and biliary excretion of calcium and strontium tracers injected into the femoral vein of dogs. They found that at 25 min after injection both isotopes were excreted into all sections of the tract, about 75 per cent of the excreted strontium and 50 per cent of the excreted calcium was found in the small intestine as compared with 20 per cent strontium and 40 per cent calcium in the large intestine. The differences found by these authors in the behavior of calcium and strontium prevent a facile extrapolation of their results to radium and emphasize the need for further experimental data if the details of the fecal excretion of radium are to be understood.

CONCLUSION

Measurements on bile fistula dogs show that the radium contributed by the bile inflow into the small intestine constitutes only a few per cent of the daily amount of radium excreted. This relative amount does not appear to change with time after a single intravenous injection of radium. Other mechanisms whereby radium could enter the gut have been considered and it is suggested that while it may be likely that secretion by the cells of the small intestine is the most important route, convincing evidence of this is lacking.

Acknowledgment—We would like to thank Professor WILLIAM B. HAWKINS, for his helpful suggestions in regard to the operative procedure and care of the biliary fistula dogs. We are indebted to Dr. DONALD COLE and to Dr. RICHARD OWINGS of the Surgery Department, for performing the bile fistula operations.

REFERENCES

1. J. B. HURSH, E. NUSSBAUM and A. LOVAAS. Unpublished data.
2. W. SALANT and G. M. MEYER, *Amer. J. Physiol.* **20,** 366, (1907).

3. J. C. Aub, R. D. Evans, D. M. Gallagher and D. M. Tibbetts, *Ann. Intern. Med.* **11,** 1443 (1938).
4. P. R. Rous and P. D. McMaster, *J. Exp. Med.* **37,** 11 (1923).
5. A. Canonico and F. C. Mann, *Surgery* **13,** 81 (1943).
6. H. P. Smith, A. H. Groth and G. H. Whipple, *J. Biol. Chem.* **80,** 659 (1928).
7. J. B. Hursh, A. Lovaas, A. Piccirilli and T. E. Putnam, *Amer. J. Physiol.* **199,** 513 (1960).
8. P. B. Hawk and O. Bergeim, *Practical Physiological Chemistry.* Blakiston, Philadelphia (1944).
9. L. Singer, M. Magsood, A. B. Medlen and C. L. Comar, *Arch. Biochem. Biophys.* **66,** 404 (1957).

Health Physics Pergamon Press 1961. Vol. 6, pp. 189–197. Printed in Northern Ireland

A NEW PROCEDURE FOR PLUTONIUM URINALYSIS*

S. MARSHALL SANDERS, JR.† and SARAH C. LEIDT

(*Received* 18 *August* 1960)

Abstract—A simple method of urinalysis, sensitive enough to detect 0.007 dis/min plutonium in 250 ml of urine, is described. In this method, anion exchange is used to separate plutonium from other inorganic ions remaining after evaporation and oxidation of the urine. The quantity of plutonium is then determined by counting α-tracks in an autoradiograph of the metal disk upon which the plutonium has been electrodeposited.

INTRODUCTION

THE initial plutonium urinalysis procedure used at the Savannah River Plant was adopted in the latter part of 1952 in anticipation of the beginning of operations. There was insufficient time to develop a unique procedure, so a study was made of existing ones. To avoid inherent disadvantages found in each method several were combined. The resulting procedure[1] contained a very sensitive technique used by The Hanford Atomic Products Operations[2], in which plutonium was first isolated by thenoyltrifluoro acetone (TTA) solvent extraction and electrodeposition and then quantitatively measured by autoradiography. The initial separation of the plutonium from urine, however, was accomplished by multiple coprecipitations with bismuth phosphate and lanthanum fluoride using a procedure developed by FARABEE[3].

Although this procedure required 1500 ml of urine and was exceedingly long and tedious, the sensitivity and precision obtained with it was acceptable and it remained in use as a routine bio-assay procedure until replaced in May 1959.

By 1956, however, the development of a new method of analyzing urine for plutonium, described in this report, had already begun.[4] The objective of this program was to develop a more sensitive but less laborious procedure than previously existed.

* The information contained in this article was developed during the course of work under Contract AT(07-2)-1 with the U.S. Atomic Energy Commission.

† Now located at the University of California, Berkeley, California.

DETAILS OF THE PROCEDURE

Evaporating and wet ashing

At the Savannah River Plant urine is normally collected during working hours in a polyethylene bottle. In the laboratory 250 ml is poured into a 500-ml Erlenmeyer flask. The bottle is then rinsed with 5 ml of concentrated nitric acid and this too is added to the flask. Forty-five milliliters of concentrated nitric acid is slowly added to the flask; the flask is swirled to mix the contents and is placed on a hot plate.

The initial acid concentration is not critical, but care must be used in adding concentrated nitric acid to avoid a rapid effervescence due to the presence of varying quantities of carbonates in the urine. (On occasion as much as 9 ml of concentrated nitric acid was needed to react with the carbonates in a single 250 ml sample producing over 800 ml of CO_2.) After the acid is added the flask is placed on $\frac{1}{8}$-in. asbestos boards on a warm hot plate. The heat is increased gradually until the sample is boiling. This gradual increase in heat prevents boiling over and ignition of the organic matter.

In practice, samples are placed on a hot plate about 4.5 hr before the end of the work day. The heat is increased twice at $\frac{1}{2}$-hr intervals. An hour is added if the evaporation is begun on a cold hot plate. At the end of the work day the hot plate is turned off, and is turned on automatically 12 hr before the beginning of the next work day. The samples, therefore, have just finished evaporating when work starts the following day. (Excessive heating of the residue causes it to become less soluble and is avoided.) At this time the flask is removed from the hot plate and allowed to cool for 5 min. Ten milliliters of concentrated nitric acid and 5 ml of 30 per cent hydrogen peroxide are added; the flask is returned to the hot plate. The solution is again evaporated and both

nitric acid and hydrogen peroxide are added repeatedly until the residue is white or very pale yellow. Ten milliliters of concentrated nitric acid is then added and evaporated almost to dryness.

Tests have shown that losses of plutonium during this operation are less than 1 per cent, even when there is vigorous boiling.

The Erlenmeyer flask used in this procedure can be re-used but should be discarded after a month's usage to prevent possible contamination build-up.

Ion exchange

Separation of plutonium from macro-quantities of other ions remaining in the residue, resulting from the evaporation and wet-ashing steps, is accomplished by using an anion exchange separation based on one described in Chalk River reports.[5–7]

The residue remaining after wet ashing is dissolved in nitric acid by gradually increasing the acid concentration. Twenty milliliters of distilled water is first added and as much of the residue dissolved as possible. Then four 5-ml portions of concentrated nitric acid are added, with gentle warming of the flask after each addition. The nitric acid concentration of the resulting solution should not exceed 8 N. This solution is allowed to stand overnight to bring all solids into solution. This rather involved procedure for dissolving the residue has been found necessary. Experience has shown that with some urine samples, smaller volumes or higher acid concentrations will cause salts to reprecipitate resulting in a loss of plutonium and clogging of the ion exchange resin.

Excessive heating and boiling is avoided as this will concentrate the acid. The capacity of the resin for plutonium is greatest when the nitric acid normality of the plutonium solution is between 7 and 8. Plutonium is believed to be in the form of a complex monovalent anion, $Pu(NO_3)_5^-$, at this normality. Even though good recoveries can be obtained from a 10 N nitric acid solution, increasing the nitric acid concentration to form the divalent complex, $Pu(NO_3)_6^{2-}$, will tend to displace plutonium from the resin due to competition between the nitrato complex and the nitrate ions for absorption.

A column is prepared for holding 0.5 ml of anion resin by placing a small glass-wool plug in the tip of a column which has a 20 mm² cross-section. The Amberlite CG-400 type 2 resin, which has been sized to remove the smaller particles, is slurried with an equal volume of distilled water and transferred to the column using a medicine dropper. After 0.5 ml of resin has been placed in the column (the height of the resin is about 2 cm), a small glass-wool plug is placed over the resin and the column is inserted in a stopper of the vacuum attachment (see Fig. 1).

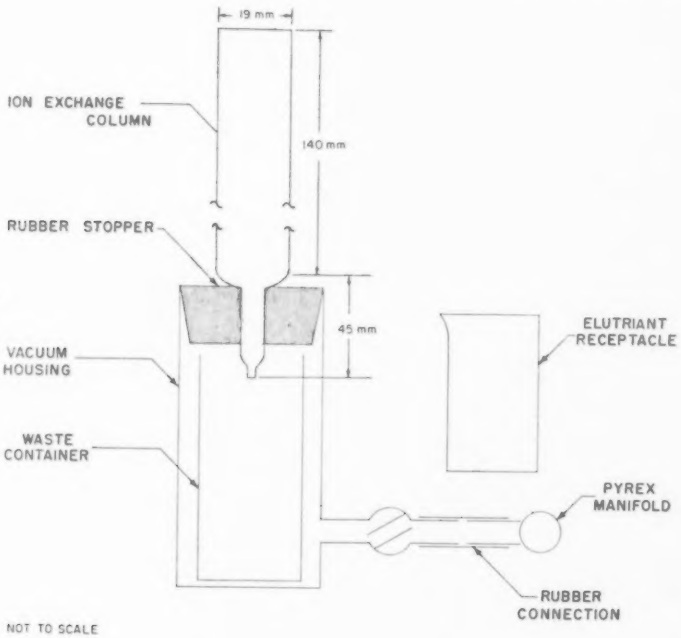


FIG. 1. Ion exchange set-up for vacuum connection.

(Resin volumes of less than 0.25 ml cause some loss of plutonium during the washing, and volumes greater than 0.5 ml require increased volumes of wash solution and eluting agent.)

A polyethylene cylinder is now placed into the main vacuum container to catch the waste solution; the stopper containing the column is fitted into the top of the container. Resin is then converted to the nitrate form by passing 1.5 ml of 8 N nitric acid through the column. The plutonium complex is then placed on the resin by passing the 8 N nitric acid solution through the column. The solution will usually pass by gravity flow. If it becomes necessary to increase the flow rate, the equipment is designed to permit regulation of the flow by the application of a vacuum (Fig. 1). This system is attached to a water aspirator or jet ejector; each column has its own regulator. The flow rate is not critical, but should not exceed 1.25 ml/min per cm^2, which is about 1 ml in 4 min from a column with a 20 mm^2 cross-section.

The resin is now washed with 8 N nitric acid to complete the removal of other ions. This is accomplished by rinsing the Erlenmeyer flask twice with 5-ml portions of 8 N nitric acid. These portions of the acid rinse are added to the columns. Concentration of the acid is then reduced by washing the resin with 2 ml of 4 N nitric acid. Plutonium begins to move off the column when it is washed with dilute acid; however, if not more than 5 ml of 1 N to 4 N nitric acid is used, less than 1 per cent of the plutonium will be lost. Washing with a solution having a nitric acid content greater than 4 N will cause gas evolution in the column when followed by sulfurous acid.

The cylinder containing the waste solutions is removed from the main vacuum container and replaced by a 20-ml beaker. Plutonium is then removed from the resin by 5 ml of concentrated sulfurous acid, due to the reduction of $Pu^{(IV)}$ to $Pu^{(III)}$, which is not absorbed on the resin. Care is taken not to allow the elution to take over 30 min.

Decontamination factors for uranium and thorium are greater than 60 for this operation.

Electrodeposition

Electrodeposition is used to prepare a thin, uniformly distributed film of plutonium for subsequent α-counting by autoradiography. The area on which the plutonium was electrodeposited in the old procedure was 7 mm in diameter. The microprojector used to count α-tracks in the emulsion could scan an area of only 4 mm $\times$ 4 mm or about one-third of the area containing tracks. The diameter of the plated area was reduced to 149 mils (3.785 mm;) this permitted all of the α-tracks to be counted and the sensitivity of the procedure to be tripled.

However, the electrolyte used previously, which was 1.6 M in potassium chloride and 1.0 M in potassium hydroxide, could not be used in conjunction with the smaller area. Recoveries resulting from a large number of alterations in both the plating conditions and electrolytic concentrations never exceeded 80 per cent and generally were much less. When a number of variations of an ammonium nitrate–nitric acid electrolyte suggested by PERKINS[8] were tried, it was discovered that reproducibly high recoveries were contingent upon the electrolyte's being made basic with potassium hydroxide just prior to removing the plating potential at the completion of the deposition. The deposition is by cathodic base formation; that is, hydrogen ions are depleted in the region of the cathode by reduction, thus causing the precipitation of plutonium-IV as a hydroxide:

$$4H_2O + 4e^- \rightarrow 2H_2 + 4OH^-$$

$$Pu^{4+} + 4OH^- \rightarrow Pu(OH)_4$$

$$Pu^{4+} + 4H_2O + 4e^- \rightarrow Pu(OH)_4 + 2H_2$$

Since the deposition is carried out in a 0.1 N nitric acid electrolyte, another cathode reaction involving the acid also occurs:

$$H^+ + NO_3^- + H_2O + 2e^- \rightarrow HNO_2 + 2OH^-$$

$$HNO_2 + 4H_2O + 6e^- \rightarrow NH_3 + 6OH^-$$

$$2Pu^{4+} + 8OH^- \rightarrow 2Pu(OH)_4$$

$$2Pu^{4+} + H^+ + NO_3^- + 5H_2O + 8e^- \rightarrow 2Pu(OH)_4 + NH_3$$

The electrolyte remains acidic throughout the deposition due to a replenishing of the hydrogen ions at the anode by the destruction of water:

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^-$$

Ammonium ions must be present in the electrolyte for deposition of plutonium to occur. They combine with the hydroxyl ions produced at the cathode to form ammonium hydroxide which is only slightly disassociated and consequently has low mobility. Therefore, a basic layer is maintained at the cathode due to the reduced mobility of the hydroxyl ions.

In preparing plutonium electrolyte for this operation sulfur dioxide should be driven off. This can be accomplished by heating or by allowing the electrolyte to remain overnight in an open container. A "Lucite" electrodeposition cell, shown in Fig. 2 is prepared by cleaning a $\frac{1}{2}$ in. diameter stainless steel disk by rinsing it in acetone, distilled water, concentrated nitric acid, and again in distilled water.

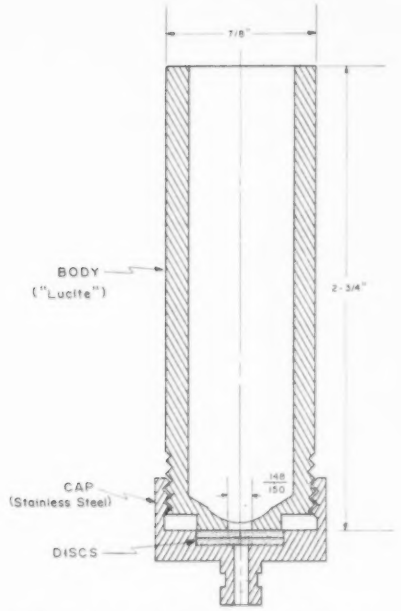


FIG. 2. Electrodeposition cell assembly.

The electrodeposition cell is then assembled by placing a second $\frac{1}{2}$ in. diameter stainless steel plate in the recess of the cell cap to serve as a shim plate. The clean stainless steel disk is then placed on top of the shim plate, and the cap is screwed on the "Lucite" cell body. The assembled cell is rinsed with 2 N nitric acid and checked for leaks by allowing distilled water to stand in it for a few minutes. One milliliter of 1.5 N ammonium hydroxide is added to the plutonium electrolyte, the cell is emptied and the electrolyte is poured into it. The 20-ml beaker which contained the electrolyte is rinsed with a number of small portions of distilled water. The rinsings are added to the cell until the total volume of the electrolyte is about 10 ml.

The cell is now placed on a twenty-sample electrodeposition apparatus.* Plutonium is deposited on the stainless steel disk by applying a potential of 15 V to the cell for 3 hr. The electrolyte is usually deficient in nitric acid at the beginning of the deposition and 2 N nitric acid has to be added dropwise to bring the plating current up to 150 mA.

After 3 hr of plating, the electrolyte is made basic by the addition of 1 ml of 12 N potassium hydroxide without interrupting the flow of current through it. The current will increase to approximately 360 mA when potassium hydroxide is added, but the cell is allowed to plate for 1 min after the addition to assure complete mixing. The cell is then emptied and rinsed with distilled water. The disk is removed and thoroughly dried under an infrared lamp.

Certain limits must be maintained on the composition of the electrolyte to obtain quantitative plutonium deposition. Ammonium nitrate concentrations of from 0.075 M to 0.3 M can be tolerated without affecting plutonium recovery. Nitric acid concentrations of from 0.1 N to 0.2 N yield the highest reproducible recoveries. Recoveries are inconsistent from electrolytes which are greater than 0.2 N in nitric acid. However, as much as 85 per cent of the plutonium has been deposited when the nitric acid concentration was 0.5 N. The recovery diminishes with increasing acidity to about 20 per cent for 1 N solutions.

Small amounts of sulfurous acid remaining in the electrolyte do not interfere with the quantitative deposition of plutonium; however, most of the sulfurous acid should be destroyed before electrodeposition.

Electrodeposition is 95 per cent complete in 2 hr, and 100 per cent complete in 3 hr. Once electrodeposition is begun, the plating potential must be maintained until the electrolyte is made basic. If the potential is removed from the cell before this time, plutonium will be redissolved by the electrolyte. About 25 per cent will be lost in 3 min and about 95 per cent in 1 hr. If the potential is interrupted, plating is continued for 3 hr after restoration. If the current drops during the course of electrodeposition, no attempt is made to increase the acidity of the electrolyte after the initial adjustment.

It should be emphasized that electrodeposition is simply a means of placing plutonium in a condition to be autoradiographed. It does not provide a means of separating plutonium from other elements, e.g. uranium, which is also deposited under the same conditions.

Autoradiography

Autoradiography is used to measure the amount of plutonium which has been electrodeposited. This portion of the procedure is similar to that previously used[1,2] except that the emulsion is treated prior to exposure to remove any latent images accumulated from contaminants in the glass or the emulsion. This is done by a method similar to that developed by PERFILOV[9,10]. The 1 in. × 3 in. Kodak nuclear track plate (type NTA) is agitated for 3 min in a chromic acid solution containing 0.2 g CrO_3 in 1 l. of distilled water. This has been found to reduce the

* Manufactured from Hanford Atomic Products Operation drawings SK-4-592 and SK-4-593.

track background by a factor of 10 over uneradicated slides (see Table 2). The slides are then washed for 8 min in running water, air dried, and placed in an autoradiographic camera (see Fig. 3).

The stainless steel disks are placed in the holes of the "Lucite" block with the plated area against the emulsion. After exposure for 10,000 min the films are developed, and the tracks within the total plated area are counted using a Leitz Model XI-C microprojector.

Calculation of results

Determination of plutonium by the new procedure gives the concentration in tracks per sample. Calculation of the plutonium concentration in a dis/min per 24-hr sample requires a conversion factor for tracks to dis/min, a volume factor for extrapolation to the 24-hr sample size, and a recovery factor. These corrections are combined into one factor for routine use as follows:

$$\frac{\text{conversion factor} \times \text{volume factor}}{\text{recovery factor}} = \text{over-all factor.}$$

A solution of 1 dis/min Pu/ml was prepared. Accurate dilutions of this solution were electrodeposited and autoradiographed and the conversion factor determined to be 3.7×10^{-4} by the following formula:

$$\text{conversion factor} = \frac{\text{dis/min per ml of solution}}{\text{number of tracks}}$$

The volume factor is merely the ratio of the 24-hr sample size, 1500 ml, to the actual volume analyzed. For a 250-ml sample this would be 6.

In calculating the plutonium concentration

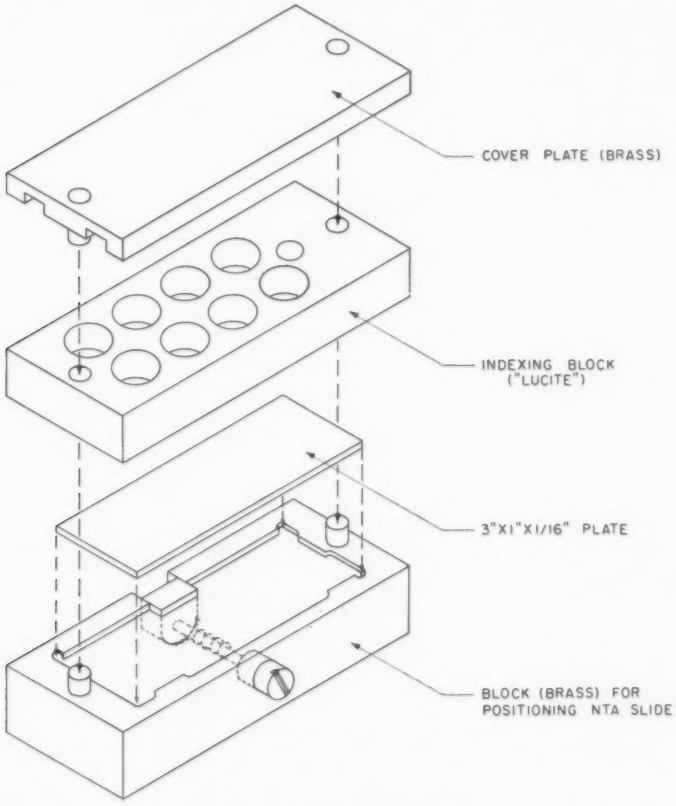


FIG. 3. Autoradiographic camera.

of the sample analyzed, it is necessary to correct the total track count for intrinsic activity in the urine. Hence, the calculation of results of the new procedure is:

$$\frac{(\text{total tracks} - \text{mean blank activity}) \times (\text{conversion factor})(\text{volume factor})}{\text{recovery}} = \text{dis/min per 1.5 l.}$$

$$\frac{(\text{total tracks} - 8)(3.7 \times 10^{-4})(6)}{\text{recovery}} = \text{dis/min per 1.5 l.}$$

EVALUATION OF THE PROCEDURE

Statistical evaluation

During a 6-week period in June and July of 1958, duplicate analyses were run with all routine samples using both the new procedure and the old or what was then the current procedure. This included blanks and spikes as well as samples from the field. Results of analyses of routine samples by both procedures are compared in Table 1. This table summarizes the analyses showing greater or less than 0.05 dis/min Pu/1.5 l. by either or both procedures.

Comparison of the two procedures was later made using forty samples collected from individuals whose urine had previously contained greater than 0.05 dis/min Pu/1.5 l. These samples, which ranged in concentration from 0.05 to 0.63 dis/min Pu/1.5 l., had an average difference between the two analyses of 0.001 dis/min Pu/1.5 l. The distribution around this mean, as shown in Fig. 4, appears to be normal. Therefore, there was no significant bias for either procedure to give a higher result than the other. There was also no relationship between the level of the analysis and the difference in the values obtained.

Table 1. Comparison of routine analyses

Results	Corrected for background*	
	Number	%
Both analyses gave <0.05 dis/min per 1.5 l.	212	64.4
Only new procedure giving >0.05 dis/min per 1.5 l.	67	20.4
Only old procedure giving >0.05 dis/min per 0.5 l.	16	4.9
Both analyses gave >0.05 dis/min per 1.5 l.	34	10.3
	Total → 329	100.0

* New procedure track value minus average track background of 2.

The limit of sensitivity for this procedure is taken to be seventeen tracks. This represents the arithmetic mean plus 2.135 times the standard deviation of the tracks from blank urine (see item 8 in Table 2). If a urinalysis results in this

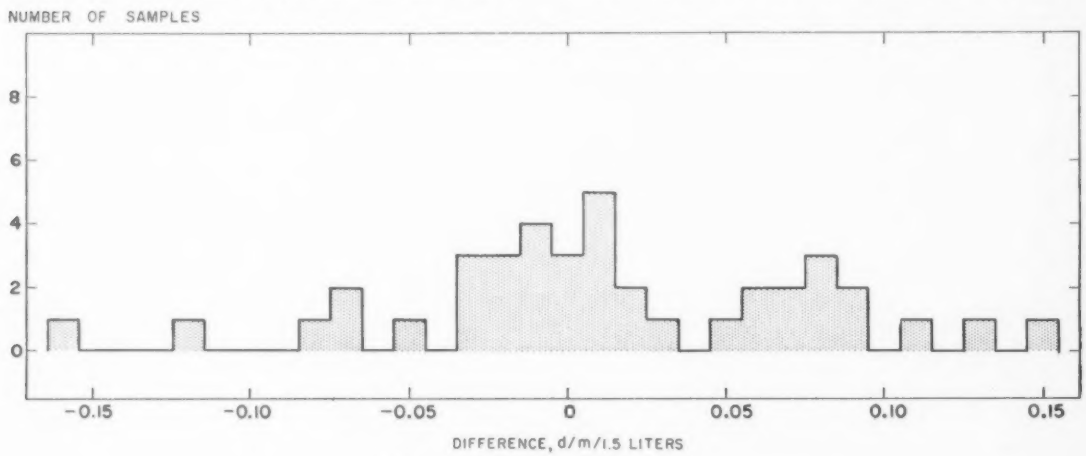


FIG. 4. Comparative differences.

Table 2. Comparison of emulsion background and blank urine analyses

	Old procedure (1/55 to 11/57)	New procedure (5/59 to 12/59)
Emulsion Background		
1. Number of determinations	1085	421
2. Arithmetic mean, tracks	27.86	2.73
3. Standard deviation, tracks	11.87	1.49
Blank Urine Including Emulsion Background		
4. Number of determinations	703	131
5. Arithmetic mean, tracks	45.57	8.34
6. Standard deviation, tracks	18.85	4.04
7. 2.135 σ, tracks	40.24	8.63
8. Mean +2.135 σ (no. 5 + no. 7), tracks	86	17
Blank Urine Only		
9. Arithmetic mean (no. 5 – no. 2), tracks	17.71	5.61
10. Estimated Pu concentration, dis/min per 1.5 l.	0.027	0.027

many tracks, odds are sixty to one against the sample containing only background plutonium. Seventeen tracks also meet an often-used criterion of being twice background. Considering the recovery at this level to be 45.4 per cent, the plutonium concentration in a urine sample producing seventeen tracks would be recorded as 0.007 dis/min per 250 ml or 0.042 dis/min per 1.5 l. Similar calculations using results obtained with the old procedure indicate that the odds were sixty to one against analysis of blank urine resulting in eighty-six or more tracks. A count of this many tracks would have been considered to represent 0.063 dis/min Pu/1.5 l. Thus with only one-sixth as much urine needed for analysis, the new procedure is 1.5 times more sensitive than the old one.

Histograms in Fig. 5 compare the distribution of the number of tracks found by using this procedure to analyze blank urine and urine with small quantities of plutonium added. Assuming these distributions to be normal, which they appear to be, urine containing 0.044 dis/min Pu/1.5 l. will produce seventeen or more tracks 52 per cent of the time, while urine containing 0.088 dis/min Pu/1.5 l. will produce seventeen or more tracks 97 per cent of the time. Comparable results were also obtained with the old procedure. Urine containing 0.061 dis/min Pu/1.5 l. would produce eighty-six or more tracks 48 per cent of the time and urine containing 0.122 dis/min Pu/1.5 l. would produce eighty-six or more tracks 96 per cent of the time.

Recoveries of the two procedures are compared in Table 3. Comparable plutonium concentrations in the new procedure are consistently lower although their precision is better. The reason for this is not known; however, the recovery appears to increase with plutonium concentration and the concentrations used in the old procedure were 1.4 times those in the new.

The standard deviation of 425 determinations of a single plutonium source, which produces 1.5 times as many tracks as the 0.146 dis/min per 250 ml plutonium spike solution, indicates that about half the total error lies in counting the tracks.

Blank urine activity

As early as 1955, an α-track count significantly above the emulsion background was observed in analyses of urine samples collected from about forty males working in the Health Physics Headquarters Building where the possibility of plutonium assimilation is nil. With the reduction in the emulsion background in the new procedure, the blank urine activity became more obvious. The arithmetic mean of the number of tracks was found to be proportional to the volume of the urine analyzed in the two procedures. This, plus the fact that different reagents and equipment were used and both procedures are specific for plutonium, might indicate that blank urine has an intrinsic plutonium concentration of about 0.027 dis/min per 1.5 l. (see item 10 on Table 2).

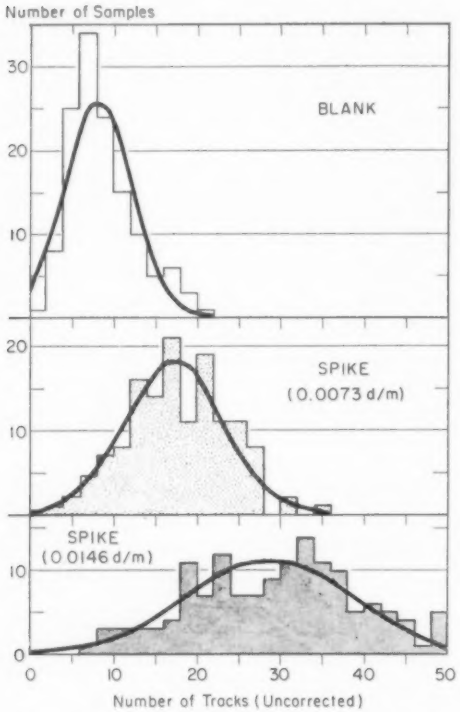


FIG. 5. Distribution of blank and spike results using the new procedure with calculated normal curves.

The possibility that this is plutonium from weapons debris is supported by data collected during 1959 by Isotopes, Inc., of Westwood, New Jersey[11] which indicates that plutonium can be detected in air, soil, various human organs and commerically available meat products. However, there is also evidence that this is not plutonium. On two occasions—November 1957 and December 1959—five distilled water samples were analyzed. In each instance four of the five analyses resulted in more tracks than the arithmetic mean for blank urine. Also tracks twice as long as the normal plutonium tracks are frequently observed in both blank urine and distilled water analyses; this indicates that at least a portion of the α-particles are from RaC′ or ThC′.

ADVANTAGES

The previous routine plutonium procedure used 1500 ml of urine (an average 24-hr void).

Table 3. Comparison of spike recoveries

Procedure used	Plutonium concentration	Number of determinations	Arithmetic mean of % recovery	Standard deviation of % recovery*
Old	0.061†	245	48.2‡	40.0
	0.122†	289	60.6‡	23.9
	1.22†	386	77.7‡	18.2
New	0.0073§	137	45.4¶	36.4
	0.0146§	142	52.0¶	23.0
	0.146§	241	63.2¶	15.3

* These data include the standard deviation of the blank urine.
† Dis/min per 1.5 l. § Dis/min per 250 ml.
‡ August 1957 to July 1958. ¶ May 1959 to December 1959.

The on-Plant sampling schedule required up to 3 weeks from the time that the sample was requested until it was received by the bio-assay group. In contrast a 250 ml sample used in the new procedure, should be received the day after it is requested. A small disposable sample container can be used to: (1) eliminate a potential source of cross-contamination, and (2) eliminate washing of sample bottles.

The time required to process a sample from the start until it was put on autoradiograph by the previous procedure was 4 days. The new procedure requires a maximum of 2 days from the beginning of evaporation until the sample is put on autoradiograph. The number of analyses made per day has been increased from 18 to 20 and the number of spikes increased from 3 to 6. At the same time, the number of technicians required to make the analyses has been reduced from 3 to 2.

Sixteen different reagents were used in the previous procedure, ten of which required preparation. The new procedure uses six reagents, four of which require a minimum of preparation time. The reduction in sample volume and the smaller equipment needed in the new procedure permit the entire operation to be carried out in only 29 per cent of the laboratory space required in the old procedure.

REFERENCES

1. S. M. Sanders, *Determination of Plutonium in Urine.* Savannah River Laboratory, DP-146, March (1956).
2. L. C. Schwendiman, J. W. Healy and D. C. Reid, *The Application of Nuclear Track Emulsions to the Analysis of Urine for Very Low Level Plutonium.* Hanford Works, HW-22680, November (1951).
3. L. B. Farabee, *Procedure for the Determination of Plutonium in Human Urine.* Clinton Laboratories, MonH-218, April (1947).
4. S. M. Sanders, *Proceedings of the Second Annual Meeting on Bio-Assay and Analytical Chemistry, October* 11 *and* 12, 1956. Los Alamos Scientific Laboratory, WASH-736, 102-105, September (1957).
5. G. M. Allison, *The Separation of Plutonium by Anion Exchange in* 10*M Nitric Acid Solution*—II. *Analytical Application to Rod Solution.* Chalk River Project, PDB-87, May (1953).
6. R. W. Durham and R. Mills, *The Absorption of Plutonium by Anion Resins*—I. *Equilibrium Studies.* Chalk River Project, CEI-62, September (1953).
7. I. W. Allam and R. W. Jones, *The Separation of Plutonium by Anion Exchange in Nitric Acid*—IV. *The Effect of Variation in Nitric Acid Concentration.* Chalk River Project, PDB-148, January (1955).
8. R. W. Perkins and D. R. Edwards, *Quarterly Progress Report, Research and Development Activities, April-June* 1954. Hanford Atomic Products Operations, HW-32406, 15–16 (1954).
9. N. A. Perfilov, *Dokl. Akad. Nauk. SSSR* **42,** 258 (1944).
10. N. A. Perfilov, *Dokl. Akad. Nauk. SSSR* **43,** 14 (1944).
11. E. P. Hardy and S. Klein, *Strontium Program Quarterly Summary Report.* Health and Safety Laboratory, HASL-77, January (1960).

Health Physics Pergamon Press 1961. Vol. 6, pp. 198–202. Printed in Northern Ireland

HEALTH PHYSICS PROBLEMS OF THE U.S. ARMY IONIZING RADIATION CENTER*

J. L. DONOVAN
Dept. of Nuclear Engineering University of Michigan, Ann Arbor, Michigan

(*Received* 7 *April* 1960)

Abstract—Some of the more unusual health physics problems associated with two very large sources of ionizing radiation are presented together with the proposed solutions. Successful solution of these problems is a vital part in the development of food processing techniques that are both safe and economical.

INTRODUCTION

Over the past decade, there have been many unique achievements in the radiation field. Radiation food preservation, the preservation of foods through the action of ionizing radiation, currently shows great promise of becoming another of the more important peaceful uses of atomic energy. The success of this seemingly novel but revolutionary method of food preservation now depends upon the effective development of an efficient production process that is both safe and economical. It is for the purpose of obtaining an efficient production process of food preservation by radiation that the U.S. Army Ionizing Radiation Center is presently being developed. This facility will be the first of its kind specifically designed and constructed to meet such an objective.

In 1953, as a result of an extensive feasibility study on the potential military application of ionizing radiation for the preservation of food, a multi-million dollar research program was initiated by the Quartermaster Corps. The objective of this research program is the development of the technology of radiation preservation of foods. This program is intended to progress to the stage at which private industry will be convinced of the commercial applications of the process.

It was realized that in order to accomplish this research objective, a pilot plant would be required. The development of the U.S. Army Ionizing Radiation Center resulted from this requirement.

The U.S. Army Ionizing Radiation Center, by the use of ionizing radiation, will provide a prototype production facility to:

(1) Determine the commercial and economic factors involved in preserving foods and other Army material.
(2) Provide process and design data which can be extrapolated to commercial production.
(3) Assist in the development of new and improved irradiation and food preparation processes.

This prototype production facility will include two different sources and types of ionizing radiation. The High Intensity Food Irradiator (HIFI) will provide γ-radiation from a multi-megacurie cobalt-60 source. The Linear Accelerator complex (LINAC) will provide high energy electrons with energies up to 32 MeV. Both units will be adjacent to a large food processing plant that will supply large quantities of food to the conveyor systems feeding each facility. A schematic layout of the U.S. Army Ionizing Radiation Center is shown in Fig. 1.

THE HIGH INTENSITY FOOD IRRADIATOR

About 2,000,000 curies of cobalt-60 will be installed in the High Intensity Food Irradiator Facility. This unit, currently under design by the Curtiss–Wright Corporation, is being built to meet the specification of the Quartermaster Corps to process 3000 lb of food per hr at a dose of 2,000,000 rads, a capacity of 3 Mrad tons/hr. A dose range of 8000 to 5,000,000 rads in a

* This work was performed under AEC contract AT(04–3)–200.

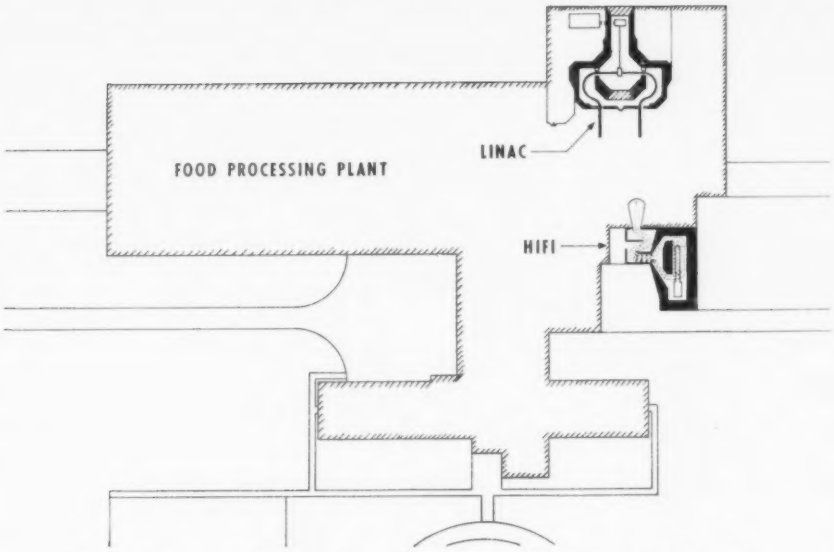


FIG. 1. Schematic layout of the U.S. Army Ionizing Radiation Center.

maximum irradiation time of 40 min is required. Variation in the conveyor speed and the provision of two conveyor systems are used to obtain this wide dose range. Dose rates inside the irradiation cell range from about 10^7 r/hr (roentgens per hour) to 10^5 r/hr. Fig. 2 shows the irradiation cell. The food packages, carried by a conveyor system, pass between the three source plaques.

LINEAR ACCELERATOR COMPLEX

The other source of ionizing radiations is a high-energy electron linear accelerator (LINAC). This unit is being designed and constructed by Varian Associates. The accelerator is capable of producing electrons with energies of from 2 to 32 MeV, and has a beam current range of from 0 to 750 μA.

The food processing capacity for the accelerator is the same as for the HIFI facility, 3 Mrad tons per hr. The food passes through the electron beam on a conveyor, and is irradiated from above and below. High-energy electrons produce a substantially uniform dose distribution in the irradiated food from the surface to a depth determined by the electron energy and the density of the irradiated food. For food packages about 6 in. in thickness irradiated from above and below, electrons with energy of about 24 MeV are required.

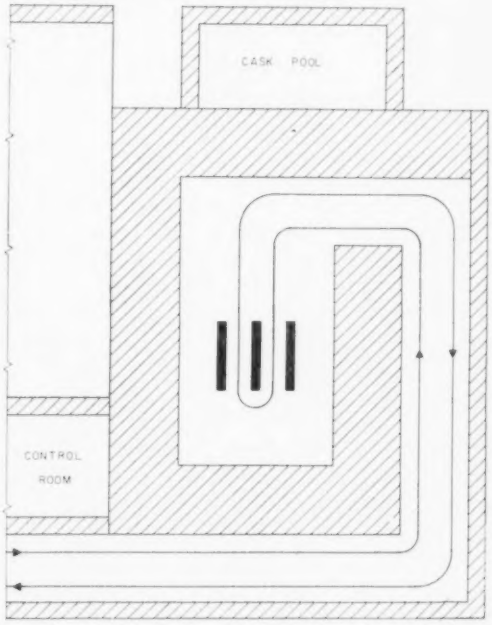


FIG. 2. High-intensity food irradiator.

HEALTH PHYSICS PROBLEMS

Both of these large energy sources have some unusual health physics problems associated with their use. For the HIFI facility, these problems involve:

(1) Shielding for 3 Mc of cobalt-60.
(2) Designing the interlocks and controls to prevent entry into the irradiation cell when the sources are in use.
(3) Design of a storage pool and lifting mechanism to raise and lower the source plaques.
(4) Providing instrumentation to detect any leak of cobalt-60.
(5) Providing area and personnel monitoring.
(6) Developing a radiation background survey program.

The shielding problem is met by the use of hollow precast concrete walls which are formed on the ground and raised into position. The concrete walls are $7\frac{3}{4}$ in. thick. Sand fill will be used between the concrete walls to provide the required shielding. The sand thickness varies from 7.0 ft to 3.0 ft. Although the original loading of cobalt-60 will probably be near 2 Mc, the shielding is designed to be adequate for a total loading of 3 Mc of cobalt-60.

Because of the high dose rates in the cell and in the corridor leading into the cell, human entry into these areas must be prevented. The food conveyors move from the loading area through the tunnel into the cell, and back to the loading–unloading area. These conveyors require two fairly large openings. To make these openings more inaccessible to human entry, they have been raised 8 ft above the floor at the entry into the tunnel.

The source storage pool has a stainless-steel liner to prevent escape of pool storage water. The source plaques are lowered to the bottom of this 20-ft deep pool when they are not in use. The pool extends under the outside wall of the cell to allow unloading of source transfer casks directly from the truck into the pool. These 15-ton shipping casks are unloaded at the bottom of the pool outside the cell, and the source plaque subassemblies are then transferred with long handling tools inside the cell for loading into the source plaques.

Each subassembly contains about 30,000 c of cobalt-60 in the form of strips 10 in. long, 1 in. wide and about 0.1 in. thick. A photograph of a representative strip is shown in Fig. 3. After irradiation at Savannah River, the strips will be removed from the irradiation can, encapsulated in welded stainless-steel subassemblies, leak tested, and then transported inside 15-ton shipping casks to the HIFI site.

To guard against a source leak after installation, monitoring systems will be installed on the pool water and air circulation systems. About 5 gal of pool water will be recirculated per min through a demineralized bed. A scintillation detector, biased to see only the cobalt-60 photopeaks, will monitor the demineralizer. For air control, a side stream of the recirculated air is to be passed through an absolute filter. A similar scintillation system will monitor the face of this filter for cobalt-60.

Monitoring is required to detect and record dose rates from 10^{-4} to 10^{7} r/hr. Dose rate monitors in the irradiation cell will record the dose rate continuously. A low-level monitor (from 0.1 to 100 mr/hr) will record the dose rate in the irradiation cell when the sources are under water. Upward movement of the sources will cause this monitor to be de-energized when the dose rate approaches 80 mr/hr. Ionization chambers are to be located in the cell to record the dose rate range of 10 mr/hr to 10^{6} r/hr. Two spherical graphite ionization chambers will monitor the dose rate range of 10^{4} to 10^{7} r/hr. The signals from these monitors are to be fed to multi-point chart recorders in the control room to provide a permanent record. One of the spherical graphite ionization chambers is to be located behind the food conveyor so as to provide a record of food processing. Low-level monitors (from 0.1 to 100 mr/hr) are to be located in the control room, in the loading and unloading area, and outside the building to provide a record of dose rates at these places. All low-level monitors have preset alarms to guard against high dose rates in these occupied areas.

Most of the problems associated with the LINAC facility are similar. One different problem results from the fact that electrons with energies higher than approximately 10 MeV will form radioactive isotopes in most elements

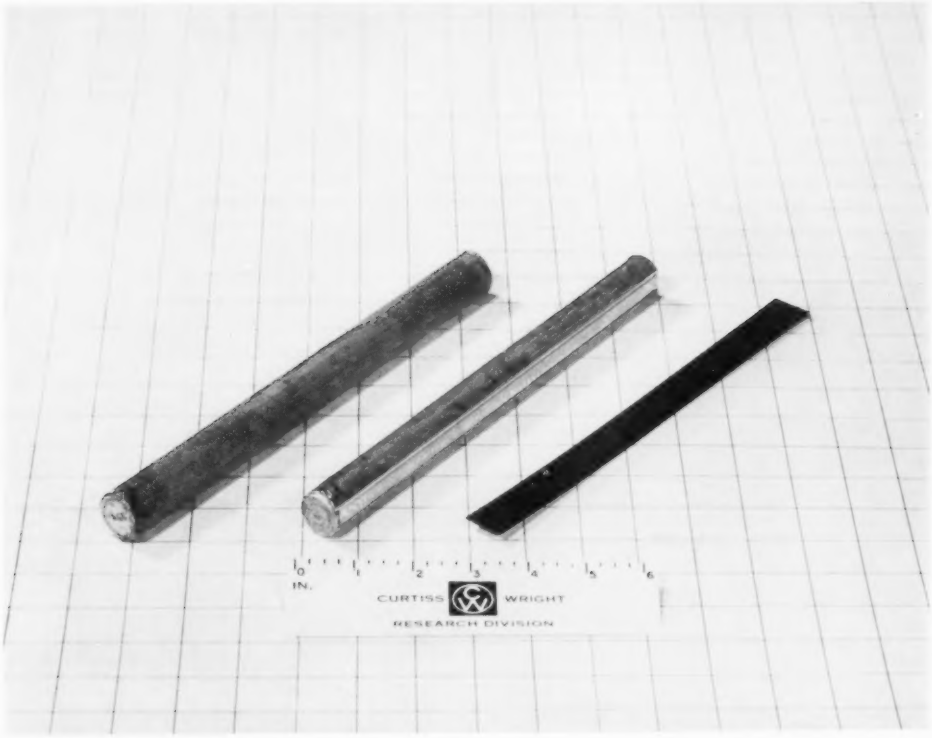


FIG. 3. Cobalt-60 strip irradiation assembly.

BLANK
PAGE

Table 1. Isotopes produced in irradiation of processed food with 25 MeV electrons

Food	Concentration in food where element is prevalent (%)	Isotope produced	Half-life	Type of decay
Many	67	C^{11}	20 min	$\beta+$
Sardines	10^{-3}	F^{18}	1.9 hr	$\beta+$
Bacon	1.2	Cl^{34}	33 min	$\beta+$
Cheese	1.6	Na^{22}	2.6 years	$\beta+$
Liver	2×10^{-3}	Cu^{64}	12.9 hr	$\beta+$, $\beta-$, E.C.
Cheese	3×10^{-4}	I^{126}	13 days	$\beta-$, E.C.
Many	10	O^{15}	2 min	$\beta+$

(Table 1). This means that, even after the beam is shut off, residual activity is present in the facility. Entry into the cell after machine shutdown will have to be controlled. Most of the isotopes formed either have short half-lives or are present only in very small quantity. However, the 20-min carbon-11 isotope and, in rare instances, the 33-min chlorine-34 isotope create a health hazard. Therefore the irradiated products cannot be handled directly by personnel immediately after processing but must be placed in storage by mechanical equipment. A storage space in which the irradiated product can be stored for approximately 5 hr must be available.

The radiation background program for the U.S. Army Ionizing Radiation Center is unusual. Whereas the conventional radiation background survey program is concerned with monitoring gross β–γ activity, this program will look for specific isotopes. The only isotope of importance to the HIFI is cobalt-60, so analysis for this isotope is a specific check for possible contamination from that facility.

Most of the isotopes formed by the γ-neutron reaction resulting from the bremsstrahlung spectrum generated by the high-energy electrons are positron emitters. These isotopes can be detected by the annihilation radiation (0.51 MeV) associated with their decay. Examples of these isotopes are sodium-22, zinc-65, carbon-11, nitrogen-13 and oxygen-15.

Analysis of the water, soil, vegetation and fallout samples collected on site and off site will be made by scintillation counting techniques. A spectrum will be prepared of count rate vs. energy, and then the spectrum will be examined for cobalt-60, sodium-22 and the position photopeak. This specific analysis will eliminate the uncertainties due to fluctuations in the gross β–γ background (Fig. 4). It is fortunate that neither cobalt-60 nor sodium-22 is a naturally occurring radioisotope.

The new values for the maximum permissible

Table 2. Permissible concentrations in air and water above natural background (continuous exposure) for cobalt-60 and sodium-22

Isotope	New values (μc/ml) Water	Air	Old values (μc/ml) Water	Air
Cobalt-60				
insoluble	3×10^{-5}	3×10^{-10}	2×10^{-2}	10^{-6}
soluble	5×10^{-5}	1×10^{-8}		
Sodium-22				
insoluble	3×10^{-5}	3×10^{-10}		
soluble	4×10^{-5}	6×10^{-9}		

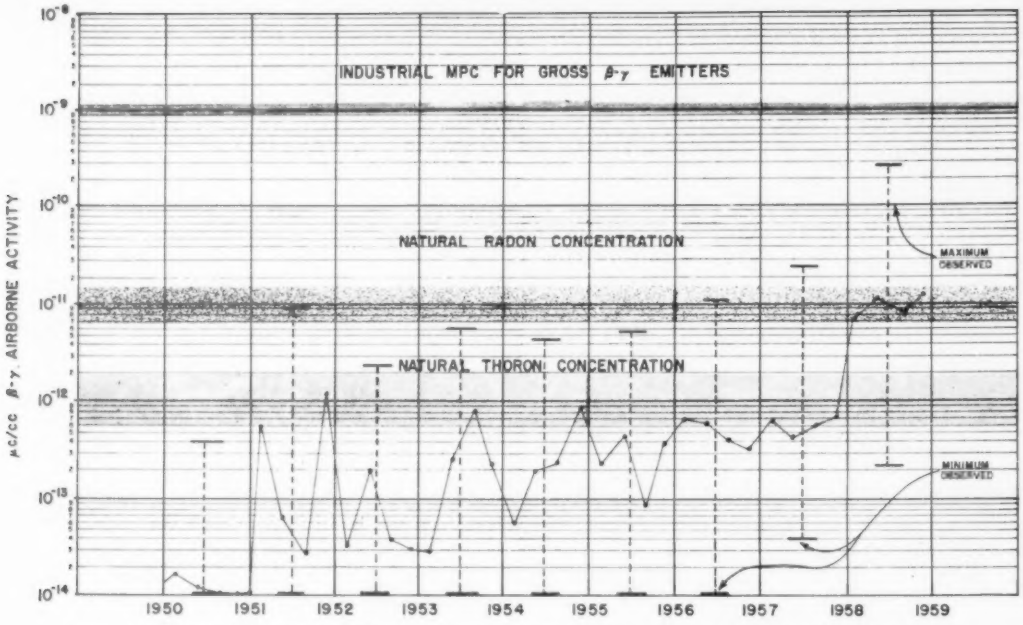


FIG. 4. Quarterly average fission products airborne activity in the San Francisco Bay area.

concentrations in air and water require that any method of analysis be more sensitive than was required by the old values (see Table 2.) For example, the values for cobalt-60 and sodium-22 shown in Table 2 require that a method of analysis should have the sensitivity to detect at least one-tenth of these concentrations.

Using a 2 in. $\times$ 2 in. sodium iodide crystal, a 1-l. soil sample, and 2 in. of lead shielding around the soil, the lower limits of detectability are 6×10^{-7} μc/ml (microcuries per milliliter) for cobalt-60, 3×10^{-7} μc/ml for sodium-22, and about 6×10^{-7} μc/ml for each if both are present. Each of these values satisfies the sensitivity requirement.

REFERENCES

1. J. OVADIA, F. HEINMENTS and A. HERSHMAN, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958 Vol. 27, Paper P/807. United Nations, New York (1958).
2. *Standards for Protection against Radiation*, Title 10 Chap. 1, Pt. 20, Atomic Energy Commission (1959).

Health Physics Pergamon Press 1961. Vol. 6, pp. 203–210. Printed in Northern Ireland

OPERATIONAL EXPERIENCE WITH KANNE IONIZATION CHAMBERS*

J. E. HOY
Savannah River Plant, E. I. du Pont de Nemours and Company, Aiken, South Carolina

(*Received* 25 *August* 1960)

Abstract—Operating experience with two types of Kanne ionization chambers is described. These chambers are used to monitor gaseous radioactive β-emitting contaminants released during the operation of heavy-water moderated reactors at the Savannah River Plant. Calibration results, γ-sensitivity, the effects of β-energy on chamber sensitivity, and the effects of such variables as temperature, pressure and humidity on the operating characteristics of these chambers are discussed.

INTRODUCTION

One of the considerations in the operation of heavy-water moderated reactors at the Savannah River Plant (SRP) is the measurement of the radioisotope, tritium. The Kanne ionization chamber has been used successfully to monitor this weak β-emitter. The Kanne is especially suitable for monitoring other equally weak emitters since their range in air is short when compared with the over-all dimensions of the chamber. Other more energetic gaseous radioisotopes are detected with reduced efficiency. This ionization chamber has been found to have wide application in the monitoring of filtered exhaust air where tritium is the preponderant gaseous radioisotope.

DISCUSSION

A block diagram of a Kanne Air Monitoring System is shown in Fig. 1. This system can either be permanently installed or mounted on a mobile cart. The system consists of an intake or sample line, filter, flowmeter, Kanne chamber, air pump and exhaust line. The current produced by ionization within the Kanne chamber is measured by a micro-microammeter and continuously recorded.

A description of the components, as shown in Fig. 1, is given in the Appendix of this report. These components are identified by SRP Instrument Equipment Piece Numbers (e.g. EP 431-AJ).

There are two sizes of Kanne chambers in use at SRP. The larger of the two, Fig. 2, is an aluminum chamber with an active volume of 51.5 l. The smaller chamber, Fig. 3, is made of stainless steel and has an active volume of 18.5 l. The principle of operation of both chambers is identical but since these two chambers do not have equal volumes, each type must be calibrated separately.

EXPERIMENTAL

Calibration of SRP chambers

The initial tritium calibration of the SRP Kanne chambers, using the 51.5-l. chamber, was done as follows:

(1) The chamber was placed in a closed circulation system as shown in Fig. 4.

(2) Air in the system was then dried by placing a desiccant (calcium chloride) in the "flask" and circulating the air overnight.

(3) The desiccant was removed and weighed.

(4) A tritiated water sample was then introduced into the system by means of a hypodermic needle. (The amount of tritiated water added was always kept below the amount of water removed by the desiccant.)

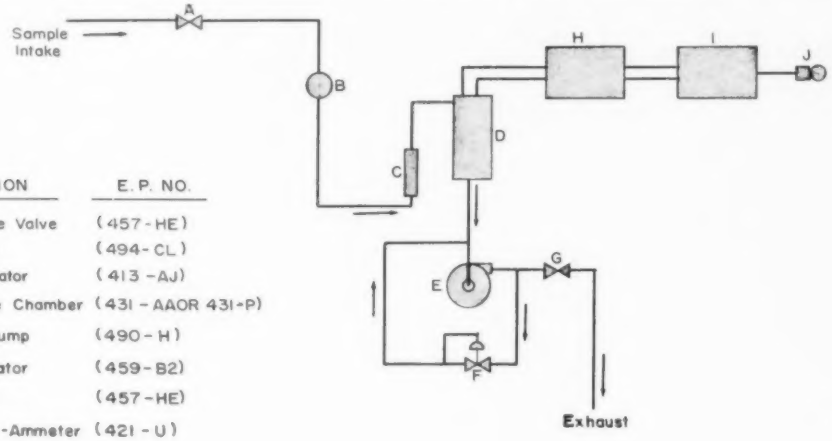
(5) After the addition of a known amount of tritiated water, the flask was heated to 125°F and air was circulated to aid in evaporation of the sample. The system was allowed to reach equilibrium before a reading was recorded.

Concentration of the tritiated water sample used in this calibration procedure was established by the

* The information contained in this article was developed during the course of work under Contract AT(07-2)-1 with the U.S. Atomic Energy Commission.

Argonne National Laboratory. No corrections were made for possible absorption losses to the walls of the system.

The average of seven experimental determinations using tritium concentrations in air varying from 2×10^{-5} $\mu c/cm^3$ to 4×10^{-5} $\mu c/cm^3$ gave an average response of 2×10^{7} $\mu c/cm^3$ per A in the 51.5-l. chamber. When the calibration of the large chamber was completed, a similar calibration was made on the 18.5-l. chamber by connecting the two chambers in series. The response of the 18.5-l. chamber to tritium was found to be 6.85×10^{7} $\mu c/cm^3$ per A. These experimental results were used as a basis for estimating the response of the Kanne chambers to any airborne



DISCRIPTION	E. P. NO.
A - Sample Valve	(457-HE)
B - Filter	(494-CL)
C - Flowrator	(413-AJ)
D - Kanne Chamber	(431-AAOR 431-P)
E - Air Pump	(490-H)
F - Regulator	(459-B2)
G - Valve	(457-HE)
H - Micro-Ammeter	(421-U)
I - Recorder	(424-BJ)
J - Alarm	(496-BS)

FIG. 1. Typical flow diagram of a Kanne air monitoring system.

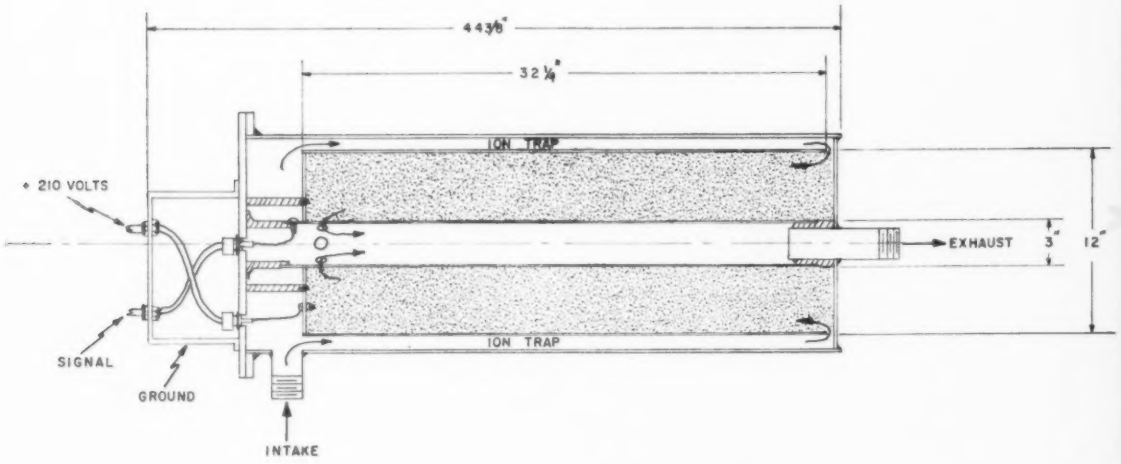


FIG. 2. Large Kanne chamber (EP-431-P).

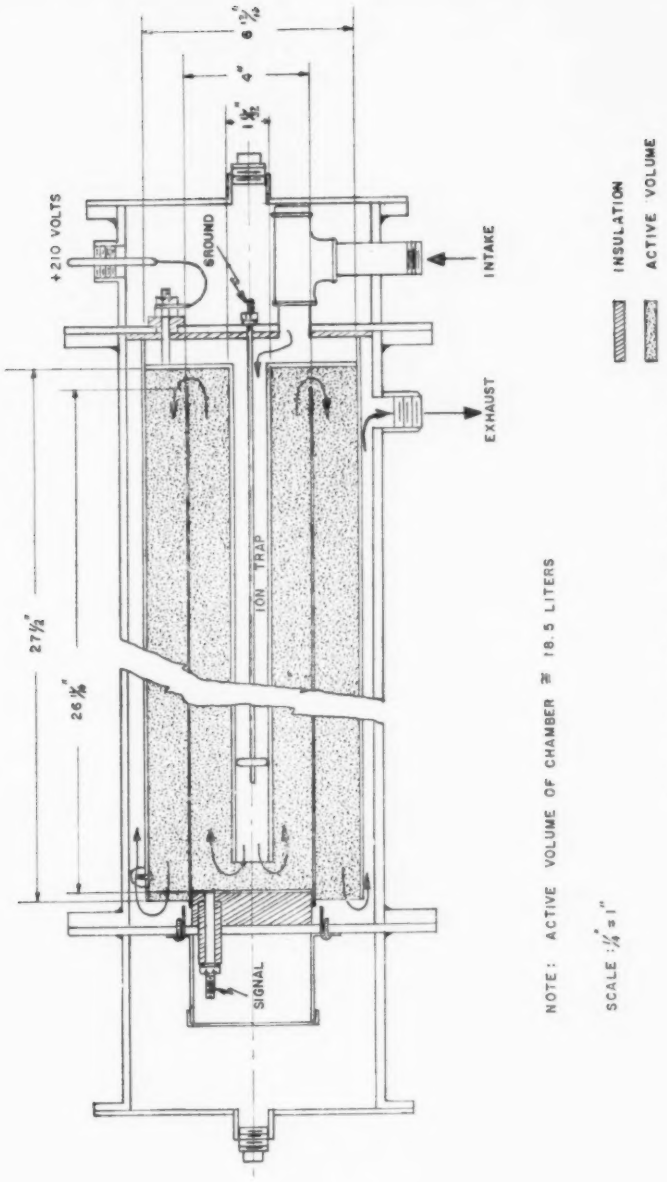


FIG. 3. Small Kanne chamber (EP-431-AA).

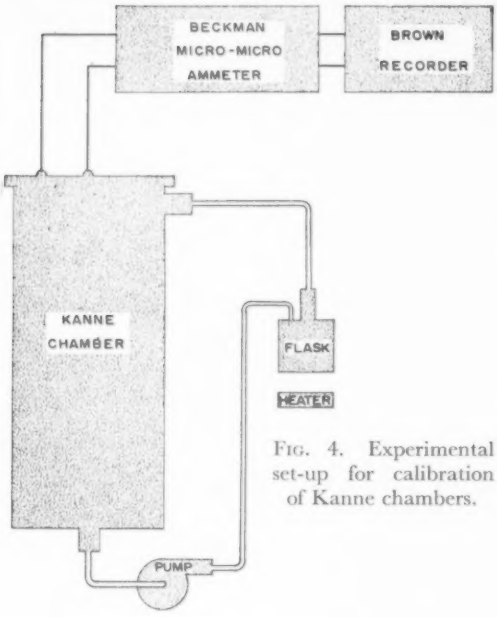


FIG. 4. Experimental set-up for calibration of Kanne chambers.

concentration of tritium. The calibration curves are shown in Fig. 5.

There is a linear relationship between the concentration of airborne activity and current as long as a sufficient voltage is present to collect all of the ions formed by radioactive decay. A study was made on the minimum voltage needed at various ion currents to maintain the chamber at "saturation". It was found that the 210 V normally supplied by the micro-microammeter was sufficient to prevent recombination of ions for currents up to 1×10^{-7} A. For radioisotopes producing ion currents greater than 1×10^{-7} A, the response is no longer linear and the calibration curves cannot be used. A collecting potential of 600 V was found to extend the linear range of the Kanne chamber to about 1×10^{-6} A.

Comparison of chamber calibrations

Calibrations of the 16.3-l. Kanne chambers at other sites have been reported[1–3]. These 16.3-l. chambers are very similar in construction to the 18.5-l. chamber described in this paper. The operating principle of the different chambers is identical; however, in order to compare calibration results,

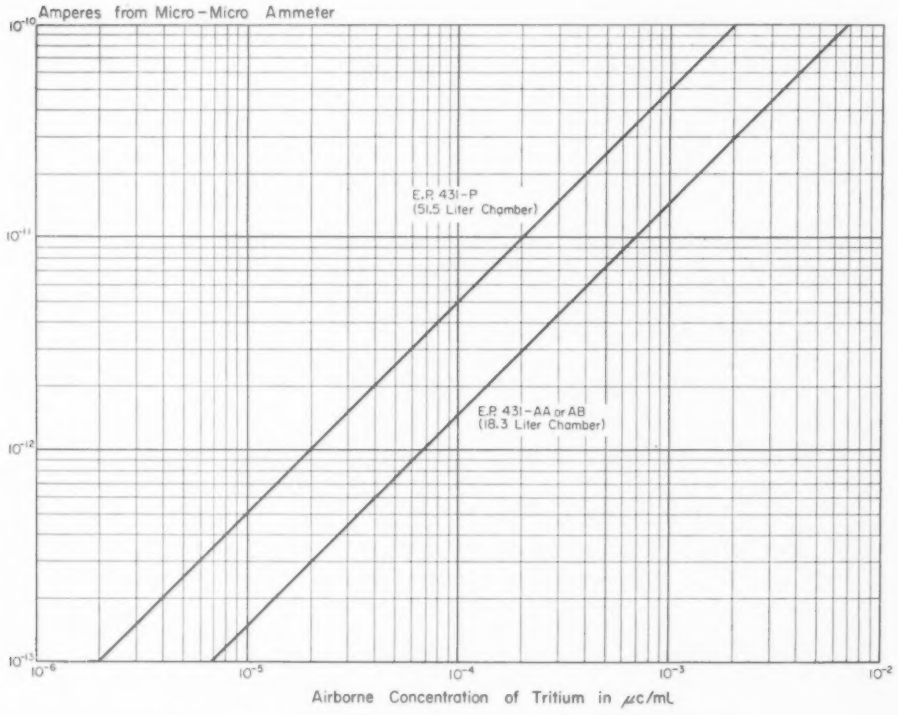


FIG. 5. Tritium calibration curve for S.R.P. Kanne chambers.

allowances must be made for their respective volumes. This was accomplished by normalizing all results to a volume of 51.5-l.

Table 1 shows a comparison between the theoretical and the experimental response to tritium for the various size chambers, all normalized to a volume of 51.5-l.

Table 1 shows that there is no disagreement in the theoretical response of the Kanne chambers if radiation losses to the walls are ignored. However, the SRP experimental results show that wall losses could cause significant error in the interpretation of data, especially from the smaller size chambers.

Table 1. Comparison of Kanne chamber calibrations for tritium

Type Kanne examined	Active volume (l.)	Calculated response* (A/μc per cm^3)	Experimental response (A/μc per cm^3)
SRP	51.5	5.65×10^{-8}	5.00×10^{-8}
SRP	18.5	5.65×10^{-8}	4.07×10^{-8}
KAPL†	16.3	5.65×10^{-8}	5.65×10^{-8}
HW‡	16.3	5.65×10^{-8}	5.38×10^{-8}

* Assuming no wall loss.
† Knolls Atomic Power Laboratory data.
‡ Hanford Works data.

Tritium loss in chamber

Theoretically, the wall losses should increase as the chamber volume is decreased; however, the apparent wall loss of 11.5 per cent in the large SRP chamber (28 per cent for the small chamber) is higher than can be justified theoretically. This loss is due in part to absorption or adsorption of tritium in and on the walls of the calibration system. No correction was made for this effect; therefore, the use of a conversion factor based on the above calibration will always lead to pessimistic answers.

OPERATING INFORMATION

Theoretically, Kanne chambers can be used to measure any gaseous β-emitting isotope dispersed in air. In practice, however, certain practical considerations limit the accuracy of such a system. Some of these considerations may be of interest to those planning such an air sampling system.

Gamma sensitivity

Calculations indicate that a γ-field of 1 mr/hr from cobalt-60 will produce 4.50×10^{-12} A in the 51.5-l. chamber and 1.61×10^{-12} A in the 18.5-l. chamber. Based on the calibration factor given in Table 1, this amount of γ-radiation would be equivalent to 9×10^{-5} μc/cm^3 of tritium in the large Kanne chamber and 11×10^{-5} μc/cm^3 of tritium in the small Kanne chamber. For this reason the chambers should be located in an area where the γ-background is low. Under ideal conditions, tritium concentrations as low as 0.5×10^{-5} μc/cm^3 can be reliably detected.

Efficiency vs. β-energy

Any ionization occurring within the "active volume" of the Kanne chamber will produce a current. The fraction of β-energy which is effective in producing current varies with β-energy. For low β-energies, almost all the energy is deposited in the chamber, with very little energy being lost to the walls of the chamber. As the β-energy increases, more of the energy is lost in the walls of the chamber.

The response of Kanne chambers for various β-emitting gases is usually calculated rather than experimentally determined. An equation was derived expressing the fraction of energy deposited in the active volume of the two SRP chambers as a function of average β-energy of the radionuclide being sampled. This equation does not account for backscatter or the increased ionization of β-particles near the end of their paths. In order to prevent the increased ionization near the end of the path from introducing an error in the calculations, certain limitations are imposed on the use of the

equation. The average β-energy (taken as one-third of the maximum energy listed for the emitter in question) must have a range in air 20 per cent longer than the longest dimension of the chamber. The shortest dimension should be at least 5 per cent of the range of the average β-energy. This equation then, only applies to average β-energies above 0.4 MeV for the 51.5-l. chamber and above 0.35 MeV for the 18.5-l. chamber. Within these limits, the fraction of the total energy emanating from the active volume of the chamber which is deposited within this volume can be calculated from the following equation:

$$F = \frac{E}{3R}\left[\frac{Z}{2} + 2\left(\frac{2a(r-a/2)^2 + 4/3\, r^3[\cos\Phi\,(\sin^2\Phi+2)]_0^\theta}{\pi(r^2-a^2)}\right)\right] \quad (1)$$

where:

F = the fraction of energy being emitted in the active volume that will be absorbed in that volume;
E = total amount of energy being emitted by radionuclides within the active volume;
R = range (in inches of air) of a β-particle, one-third of the maximum energy;
Z = axial length of the active volume of the chamber;
r = outer radius of the active volume of the chamber;
a = inner radius of the active volume of the chamber (outer radius of the central electrode);
θ = arc cos a/r.

Solving the above equation for the two types of Kanne chambers in use at SRP:

$$F = \frac{24.9}{3R} \text{ for the 51.5-l. chamber} \quad (2)$$

and

$$F = \frac{15.8}{3R} \text{ for the 18.5-l. chamber} \quad (3)$$

The general calculation of current response of a Kanne chamber is as follows:

$$\text{A}/\mu\text{c per cm}^3 = \frac{(3.7 \times 10^4 \text{ dis/sec per } \mu\text{c per cm}^3)\ (\text{volume in cm}^3)\ (E \text{ avg. in eV}/d)F}{(6.24 \times 10^{18} \text{ ip/sec per A})\ (\text{W in eV/ip})}$$

where:

W = energy necessary to create an ion pair;
ip = ion pair;
dis = disintegrations.

Using 32.5 for W:

$$\text{A}/\mu\text{c per cm}^3 = 9.40 \times 10^{-12}(E \text{ avg eV/dis})(F) \text{ for the 51.5-l. chamber,} \quad (5)$$

and

$$\text{A}/\mu\text{c per cm}^3 = 3.43 \times 10^{-12}(E \text{ avg eV/dis})(F) \text{ for the 18.5-l. chamber} \quad (6)$$

An investigation was made of the manner in which F varies with average β-energy as indicated in equations (2) and (3). These results are shown in Fig. 6. The dotted portion of the curve represents β-energies, below which equation (1) is not applicable. The curve has been extrapolated to experimentally determined points obtained using tritium. The fraction of β-energy deposited in the chamber is found to decrease rapidly with increased β-energy.

When the range of the average β-energy exceeds the longest dimension of the chamber, the fraction of energy being deposited does not change as rapidly as before. Fig. 7 shows the effect that the changing efficiency has on Kanne chamber response as predicted from equations (5) and (6). From these curves, an estimation of the Kanne response for any β-emitting gas can be made. Obviously, mixtures of β-emitting gases cannot be interpreted unless the relative abundance of each isotope is known.

Effects of pressure variations

Normal variations of atmospheric pressures are quite small compared to the variations which are introduced by the arrangement of an air pump and filter. The usual position of the pump, Fig. 1, is downstream from the chamber. In this position the Kanne chamber is operated at pressures slightly less than atmospheric due to the pressure drop developed by air flowing through the filter. The maximum variation of pressure drop across the filter is estimated to be less than 10 per cent of the existing atmospheric

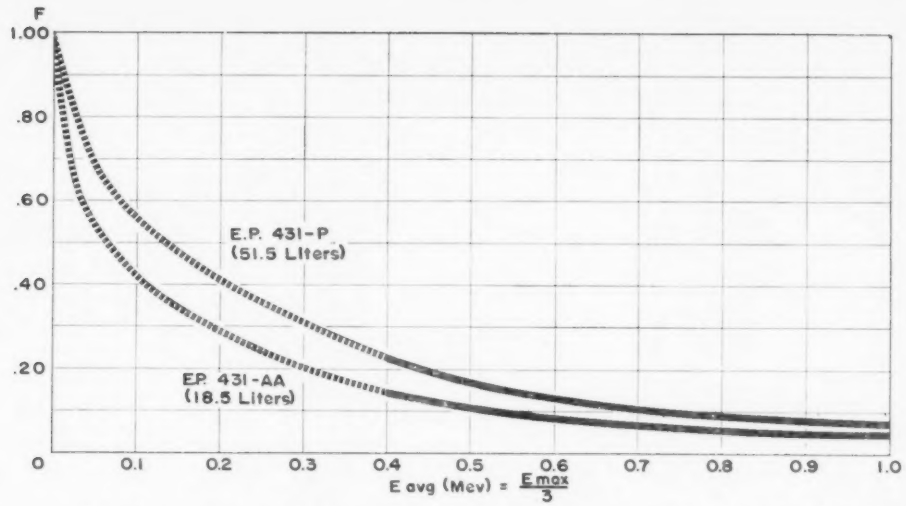


FIG. 6. Fraction (F) of β-energy deposited in the Kanne chamber vs. Function of average β-energy.

pressure and therefore well within the accuracy of the calibration. No corrections are made for changes in response due to pressure or temperature variations.

Changes in flow rate cause temporary currents which can disrupt normal readings for several minutes after they have occurred. With the exception of the initial start-up or shutdown, flow rate through a chamber should be well regulated to avoid these transient currents. In general, the choice of flow rate through the system should be governed by a compromise of the following effects.

(1) At low flow rates, the ion trap is more efficient for the removal of unwanted ions.

(2) It is desirable to operate the Kanne chamber as close to atmospheric pressure as possible. The pressure drops across the filter will vary directly with the flow rate.

(3) The delay time between sampling and reading is a function of the length of the sample line and the flow rate of the air in this line. A flow rate should be selected which is sufficiently high to avoid lengthy delays between sampling and detection.

At SRP the flow rates of the Kanne systems are set between 2 and 4 ft^3/min.

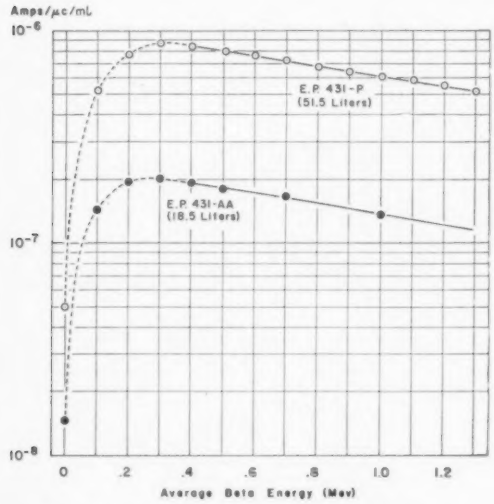


FIG. 7. Predicted change in Kanne chamber response as a function of average β-energy.

Effects of humidity

Variations in the relative humidity up to 96 per cent have been found to have no effect on the Kanne reading. In practice, these units have been operated successfully in atmospheres approaching 100 per cent relative humidity. Condensate in the chamber will disrupt the

reading by shorting the central electrode to ground or to the high voltage electrodes. Where extremely moist air streams are sampled constantly, heating elements have been installed on the exterior surface of the chamber to prevent condensation within.

Contamination problems

Experience has shown that very few contamination problems exist if the air is filtered. High concentrations of some radiogases, however, have a tendency to build-up residual activity which may require several hours of air purging to remove. Dry air and heat have been utilized successfully to speed decontamination when the contaminant is gaseous. The chamber has been constructed so that it can be disassembled for decontamination when necessary.

SUMMARY

A typical Kanne Air Monitoring System is described. Theoretical and experimental calibration of two types of Kanne chambers are compared with similar studies made at other sites. Some of the variables and limitations of such a system are discussed.

APPENDIX

Equipment piece numbers	Description
413–AJ	Indicating rotameter with a range from 0.5 to 5 ft^3/min of air
421–U	Vibrating reed electrometer with logarithmic scale to cover current range from 10^{-13} to 10^{-7} A
424–BJ	Single-point strip-chart recording potentiometer with a six-decade logarithmic scale
431–P	Ionization chamber (large Kanne) with ion collecting volume equal to 51.5-l.
431–AA	Ionization chamber (small Kanne) with ion collecting volume equal to 18.5-l.
457–HE	Hand-operated butterfly valve
459–B2	Air relief valve; flow, 4 ft^3/min of air at pressures up to 15 in. water gage
490–H	Air pump with 4 ft^3/min capacity at pressure differential of 2 in. water
494–CL	Air filter capable of filtering particles larger than 2 μ
496–BS	Alarm bell

REFERENCES

1. J. J. Fitzgerald and B. W. Borelli, *Determination of Efficiency of Kanne Chambers for the Detection of Radiogases.* KAPL-1231, Secret (1954).
2. G. E. McCullough, *Kanne Chamber Calibration.* HW-23921, Secret (1952).
3. P. S. Eisencher, *Calibration of Ion Chamber.* HW-18780, Secret (1950).

Health Physics Pergamon Press 1961. Vol. 6, pp. 211–225. Printed in Northern Ireland

NOTES

Measurements of Fast Neutron Flux by Means of Activation of Phosphorus

(*Received* 22 *March* 1961)

REINHARDT and DAVIS have described[1] a simple method for the concentration of P^{32} in sulphur irradiated with fast neutrons. The method consists of burning out the sulphur in a flat tray and it is especially recommended for the measurement of very low fluxes of fast neutrons. It has recently been pointed out[2] that the reaction P^{31} (n,p) Si^{31} can be used to measure neutrons of the same energy as those of the reaction S^{32} $(n,p)P^{32}$, but that the former method is much more sensitive if one is dealing with short irradiations. In the same paper, a method is described for the measurement of the activity in 20 g of phosphorus, but this method is not suitable for anti-coincidence counters.

A chemical separation of Si^{31}, described in the same paper, acts only with fairly small quantities of phosphorus irradiated as phosphate.

For this reason we have developed a procedure for the concentration of Si^{31} in phosphorus, analogous to the treatment of sulphur mentioned in the paper by REINHARDT and DAVIS. It was found satisfactory to evaporate about 20 g of red phosphorus from a large-size porcelain boat in a current of nitrogen and to burn the phosphorus vapour in the flame of a bunsen burner. It is convenient to have the porcelain boat in a porcelain tube heated by means of an electric furnace and to have Pyrex tubing sealed on to each end of the porcelain tube.

During the evaporation of the phosphorus, the temperature is raised from about 500 to about 600°C and, after the phosphorus has disappeared, the temperature is raised to 750 or 800°C, depending on the kind of phosphorus used. At this stage the ash is left to cool and the weight of the ash in the boat is determined. Now as much of the ash as can be scraped out of the boat is collected in a measuring tray and the activity is measured under a β-counter. An anti-coincidence counter (Philips No. 18515 & 18517) is especially useful if low neutron fluxes are to be measured. The decay of the sample is measured and a correction is applied for the incomplete transference of ash.

Four different types of commercial red phosphorus have been used to test the completeness of the retention of the Si^{31} activity in the ash. Within the accuracy of our measurements (maximum deviation about 15 per cent), the total Si^{31} activity was found in the ash.

The sensitivity of the method depends largely on the ash-content of the phosphorus, as a smaller ash-content makes it possible to work with a larger quantity. Owing to self-absorption, it is useless to measure the activity of too large a quantity of ash. In our experience the ash-content of commercially available red phosphorus amounts to between 0.7 and 3 per cent. In the case of one of the products with which we have worked the ash tended to melt, which made it impossible to collect it from the porcelain boat.

A complication is presented by the fact that red phosphorus always contains a measurable quantity of radioactivity. This contamination can be recognized by continuing the activity measurements after the Si^{31} has decayed. This activity contamination was found to amount to between 0.3 and 1.5 dis/min per g of phosphorus.

If the best quality phosphorus is used it is possible to detect a continuous flux of fast neutrons ($E >$ 2 MeV) around 20 n/cm²-sec or a single burst around 3×10^5 n/cm². This method has the advantages of being equally sensitive to pulsed neutrons and to a constant flux and of not being disturbed by the presence of γ-rays.

Acknowledgements—This investigation is part of the programme of the Foundation for Fundamental Research of Matter (F.O.M.). It has been carried out with the financial support of the Netherlands Organization for Pure Scientific Research (Z.W.O.). We wish to thank the cyclotron crew for a large number of irradiations and Mr. L. DE GALAN, Mr. R. BAC, Mr. TH. A. WEBER, Mr. B. UMANS, Mr. W. DAVIDS and Mr. P. W. KUIT for preliminary measurements.

A. H. W. ATEN, JR.
E. J. DE HAAN
W. M. C. DE JONG

Instituut voor Kernphysisch Onderzoek
Amsterdam
Netherlands

References

1. P. W. REINHARDT and F. J. DAVIS, *Health Phys.* **1,** 169 (1958).
2. D. RICABARRA, R. TURJANSKI and A. H. W. ATEN, JR., *J. Nucl. Energy* A **12,** 177 (1960).

A Broad-Range Chemical Dosimeter

(*Received* 15 *March* 1961)

I. Introduction

CHEMICAL dosimeters have been used extensively to measure gamma dose.[1] They possess numerous advantages over other types of dosimeters, especially in the measurement of absorbed dose. Unfortunately, many of these systems possess the disadvantage that rather high doses are required before the radiolysis products can be precisely measured. To overcome this, a number of chemical dosimeters exist whose fundamental reaction is a chain process. A chain reaction, although it makes the dosimeter sensitive, also generally creates a dependence of yield on the rate at which the radiation is delivered to the system. Among chemical dosimeters, a number exist where the major component of the system is water.[2] Such systems have the advantage of energy-absorption characteristics that are fairly similar to biological systems and, hence, may be used in biological dosimetry studies. The use of ferrous sulfate and ceric sulfate dosimeters has been extensively documented and, in fact, the radiolysis of water has been studied more extensively than any other chemical compound.

In devising a chemical dosimeter with biological applications, one would desire a highly sensitive aqueous system that does not utilize a chain reaction. This system should also be rather simple to prepare and be stable to its surroundings. In light of these qualifications, we have chosen to study saturated aqueous solutions of benzene as a broad-range chemical dosimeter. The radiolysis of aqueous benzene has been studied by a number of workers in the past[3–9] and the characteristics of this system are fairly well known. A major radiolysis product in the presence of air is phenol. PHUNG and BURTON[7] report a yield for phenol in the aerated system of 2.64 molecules of phenol per 100 eV absorbed. This value is in agreement with the results of investigation by other workers and is reported to be independent of dose over the range 0–3.5 $\times$ 10^4 r. A mechanism has been proposed for the radiolysis of aqueous benzene which shows no chain reaction in the irradiated system.[7]

The methods of phenol analysis, previously used in the radiolysis work, have a lower limit of 3.3 $\times$ 10^{-5} molar phenol. This is comparable to a minimum dose of 8.2 $\times$ 10^3 r.

The literature contains many references to the analysis of phenol in micro-molar quantities.[10–14] The incorporation of a highly sensitive analytical procedure for the determination of phenol with the radiolysis of benzene should create a chemical dosimeter capable of accurately measuring dose over a broad range. The procedure chosen for phenol analysis, first described by GIBBS,[11] involves the combination of phenol with 2,6-dibromo-n-chlorobenzoquinoneimine to produce the intensely blue 2,6-dibromo-indophenol. This reaction, described more completely by ETTINGER and RUCKHOFT,[12] is routinely used for phenol analysis. Since phenol is detectable in the 10^{-8} molar concentration range by this procedure, aqueous benzene could be used to measure accurately doses as low as 10 r.

II. Procedure

A. *Reagents*

1. *Benzene.* ACS Grade thiophene-free benzene (Baker & Adamson) was purified by distillation through a 30-in. Widmer column.

2. *Water.* Conductivity water prepared by passage through a mixed-bed ion-exchange column followed by double distillation in an all-quartz apparatus.

3. *2,6-dibromo-n-chlorobenzoquinoneimine* (Eastman 2304) 0.2 g reagent dissolved in 50 ml ethanol and used as a stock solution. 7.5 ml of this stock solution is diluted to 100 ml with water immediately prior to use.

4. *n-Butyl alcohol.* Fisher Certified Reagent

B. *Dosimeter system*

Dosimeter solution. Air-saturated conductivity water is saturated with benzene by vigorous stirring in an open container for $\frac{1}{2}$ hr. The solution is then permitted to stand until all excess benzene has separated from the aqueous phase. The aqueous phase then is placed in volumetric flasks and stoppered so as to leave no air space.

Dosimetry. A 100-c Co^{60} source used for the irradiation was calibrated with ferrous sulfate by the standard procedure[15] using a G of 15.6 ions per 100 eV absorbed.

Phenol analysis. After irradiation a 30-ml sample of the benzene solution was placed in a flask to which were added 1.5 ml of pH 9.5 buffer and 0.5 ml quinoneimine reagent. The solution was shaken and permitted to stand for 24 hr. After standing, 7.5 ml n-butyl alcohol was added. The n-butyl alcohol phase was extracted and its absorbance at 650 mμ was measured on a spectrophotometer (Perkin–Elmer Model 4000A Spectracord). Suitable blanks of unirradiated aqueous benzene were used. Phenol concentrations were obtained from a calibration curve prepared with known concentrations of phenol.

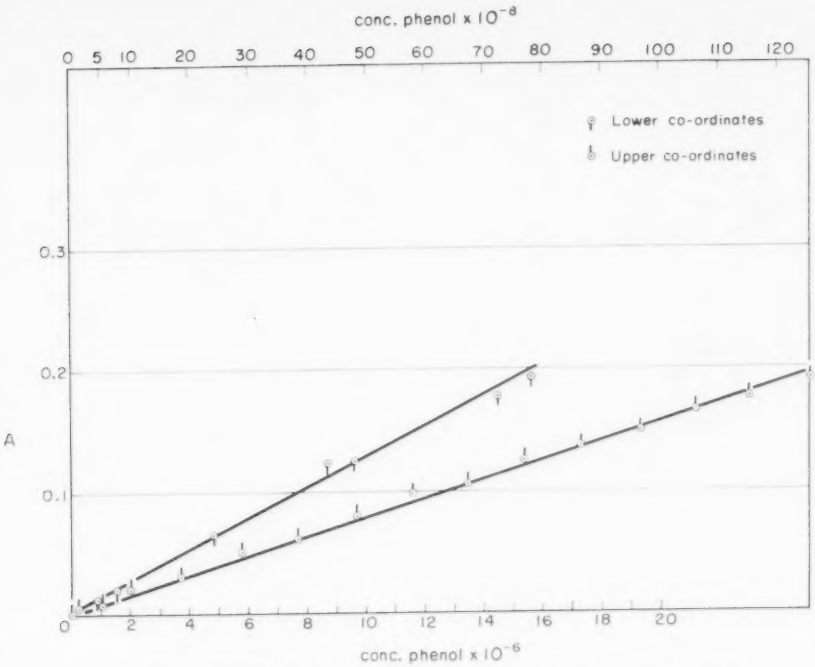


FIG. 1. Absorbance at 595 mμ (lower co-ordinates) and 650 mμ (upper co-ordinates) as a function of phenol concentration.

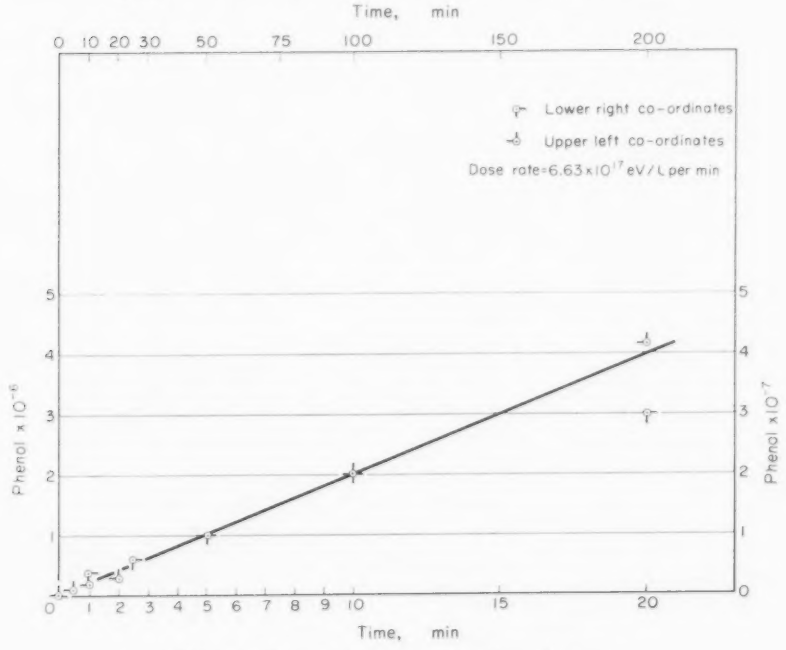

Time, min
0 10 20 30 50 75 100 150 200
Lower right co-ordinates
Upper left co-ordinates
Dose rate=6.63x10^{17} eV/L per min
Phenol x 10^{-6}
Phenol x 10^{-7}
0 1 2 3 4 5 6 7 8 9 10 15 20
Time, min

FIG. 2. Phenol yield as a function of dose.

III. Results

A typical calibration curve for the phenol analysis is shown in Fig. 1. In the event that the phenol concentration is as high as 10^{-6} molar, the butyl alcohol extraction can be eliminated and the indophenol absorption read directly on the spectrophotometer. The absorption maximum for the aqueous solution is 595 mμ. Analysis in the range from 10^{-6} molar to 10^{-4} molar is accomplished in this way. Above 10^{-4} molar, the sample must be diluted. Typical results of the radiolysis are shown in Fig. 2. It can readily be seen that the sensitivity of the system is quite good. Conventional care in the preparation of the benzene solutions and in conducting the analysis permits a measurement of dose with a precision of ±10 per cent.

Acknowledgment—The author wishes to thank Mrs. RETA SHERMAN who tirelessly performed the large number of analyses necessary for the completion of this work.

NATHAN KLEIN

U.S. Army Chemical Corps
Nuclear Defense Laboratory
Army Chemical Center, Maryland

References

1. G. V. TAPLIN, *Radiation Dosimetry* (Edited by G. J. Hine and G. L. Brownell) p. 358. Academic Press, New York, (1956).
2. E. J. HART, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958, P/29, p. 5. United Nations, New York (1958).
3. G. STEIN and J. WEISS, *J. Chem. Soc.* 3245 (1949).
4. T. J. SWORSKI, *J. Chem. Phys.* **20,** 1817 (1952).
5. T. J. SWORSKI, *Rad. Res.* **1,** 231 (1954).
6. J. H. BAXENDALE and D. SMITHIES, *J. Chem. Phys.* **33,** 604 (1955).
7. P. V. PHUNG and M. BURTON, *Rad. Res.* **7,** 199 (1957).
8. J. GOODMAN and J. STEIGMAN, *J. Phys. Chem.* **62,** 1020 (1958).
9. K. C. KURIEN, P. V. PHUNG and M. BURTON, *Rad. Res.* **11,** 283 (1959).
10. H. D. GIBBS, *J. Biol. Chem.* **71,** 445 (1927).
11. H. D. GIBBS, *J. Biol. Chem.* **72,** 649 (1927).
12. M. B. ETTINGER and C. C. RUCKHOFT, *Analyt. Chem.* **20,** 1191 (1948).
13. M. B. ETTINGER, C. C. RUCKHOFT and R. J. LISHNA, *Analyt. Chem.* **23,** 1783 (1951).
14. L. J. SCHMAUCH and H. M. GRUBB, *Analyt. Chem.* **26,** 308 (1954).
15. R. H. SHULER and A. O. ALLEN, *J. Chem. Phys.* **24,** 56 (1956).

Pre-Operational Health Physics Training of Reactor Personnel

(*Received* 29 *September* 1959)

Preface

A. *Purpose of note*

To present a reactor health physics training program as an operational problem.

B. *The health physicist's job*

(1) Set up a sound radiological safety training program based on operational needs as he sees them.
(2) Sell the training program to management and obtain managerial support.
(3) Present the program and make it fulfill its purposes. Be flexible. (Recognize the needs of janitors as well as engineers in the presentation.)

Practicable Health Physics Indoctrination Lecture

A. *Introduction*

Purpose of lecture:

(1) To inform personnel of the problems in working with radiation and radioactive materials.
(2) To inform personnel of proper procedures to be followed for radiation safety.

B. *Basic concepts*

(1) Nuclear radiation and the induction of biological effects.
  (a) Energy deposited in body from radiation may cause biological effects (medical injury).
  (b) Sunburn analogy.
  (c) Radiation protection achieved by controlling the amount (dose) of radiation received.

(2) Biological effects of radiation (medical injury). Reference: *Radiation Safety Manual*, Chap. VI.
  (a) Conditions of exposure:
    (i) external—irradiation from a distance;
    (ii) internal—radioactive material enters body through breathing, eating, wounds, etc.;
    (iii) acute—sudden exposure to large dose;
    (iv) chronic—long continued exposure to low doses.
  (b) How much exposure to allow?—based on acceptable risk.

C. *Radiation exposure limits—measurement and control*

(1) Maximum Permissible Exposure Limits 100 mrems/week averaged over 1 year, not to exceed 300 mrems in any 1 week.

(2) Overexposures.
  (a) Technical overexposure—300-900 mrems in 1 week (allowable under penalty with health physics permission).
  (b) Overexposures—more than 900 mrems in 1 week.

(3) Measurement of dose.
  (a) External—film badges, dosimeters, pocket chambers.
  (b) Internal—bioassay, urinalysis.

(4) Control of exposure.
  (a) External—time, distance, shielding.
  (b) Internal–contamination control use of respirator and masks, protective clothing and equipment.
  (c) Health physics surveillance and control of radiation work

(5) Penalty for overexposure—removal from radiation work may mean loss of job.

D. *Radiation exposure control measures*

(1) Procedures for use of film badges, dosimeters, pocket chambers (Prototype Instruction J/20/50-58) obtained originally from Health Physics and safety. Film badges and dosimeters picked up and returned daily to guard station. Pocket chambers obtained for special work from health physics.

(2) Training of personnel in radiation safety.
  (a) Practical indoctrination lecture.
  (b) Special training.

(3) Controlled areas and clean areas.
Isolation of radiation zones and control of work, traffic and the movement of materials.
Controlled areas in Building Nos. 6, 5, 5A and Hull.

(4) Use of protective clothing and equipment.
Clothing and equipment for use in controlled areas is available from Health Physics.
Includes—lab. coats, coveralls, head covers, gloves, shoe covers, rubbers, respirators, masks, gauntlets, goggles.
Purpose—to prevent contamination of body surfaces, inhalation or ingestion, spreading of contamination.

(5) Control of contamination. (Contamination—loose radioactive material, dusts, liquids, powders etc.)
  (a) Control stations—signs, ropes, protective clothing and equipment.
  (b) No eating, smoking, drinking.
  (c) Use of locker, change room, showers, wash basins.
  (d) Use of radiation monitoring equipment.
  (e) Decontamination of equipment and materials.
  (f) Waste disposal: (i) liquid-waste system; (ii) solid-waste procedures.

(6) Operation and functions of the Health Physics and Safety Group
  (a) Control of radiation work and work conditions.
  (b) Radiation exposure control: (i) exposure records; (ii) radiation surveys and monitoring; (iii) assignment of working times.
  (c) Technical assistance to supervisors.
  (d) Establishment of safe working practices.
  (e) H.P. approval for radiation work.
  (f) H.P. instructions for radiation work.
  (g) Training of personnel in radiation safety.
  (h) General safety work.

(7) Rules for shipment, storage and transportation of radioactive materials.
  (a) ICC regulations requirements for packaging and labeling of shipments, classes, radiation limits.
  (b) Prototype procedures—monitoring and record keeping.

(8) Responsibility for visitors, outside contractors etc.
Person responsible must obtain film badges, dosimeter protective clothing and equipment and see that visitors follow established procedures.

E. *General safety*

Work involves:

(1) Safe operation of machinery and equipment.
(2) Establishment of safe working conditions and safe working practices.
(3) Issuance of safety equipment as goggles, shoes, gloves etc.

F. *Outline of radiation safety responsibilities.*

G. *Question and answer period*

Practical Health Physics Training for Supervisors

Pre-requisite

Attend Practical Health Physics Indoctrination lecture.

Text

Radiation Safety Manual SlC-0021-TM-6.

Session I–Subjects and topics for discussion

(1) Radiation control.
(2) Safe handling of radioactive material.
(3) Biological effects of radiation.
(4) Permissible exposure.
(5) (a) Health Physics approval for radiation work.
(b) Health Physics instructions.
(6) Issuance of film badges and generation of required records and training of personnel in radiation safety. Also Prototype Instruction J20/No. 50-58 and Prototype Instruction J20/No. 51-58.
(7) Controlled, uncontrolled and clean areas.
(8) Control of contamination.
(9) Question of period.

Session II—Subjects and topics for discussion

(10) Decontamination.
(11) Protective clothing procedure.
(12) Health physics survey and monitoring activities.
(13) General safety activities.
(14) Operation radiological exposure problems and control of radiation exposure.
(15) Individual responsibilities for waste disposal.
(16) Rules for the procurement, receiving, shipping and transportation of radioactive materials.
(17) Emergency procedures.
(18) Question period.

L. Lewis

Combustion Engineering, Inc.
Naval Reactors Division
Windsor, Conn.

Instruments for Civil Defense

(*Received* 4 *December* 1959)

The Office of Civil and Defense Mobilization has for several years been actively engaged in the development and purchase of radiological instruments for the use of organized civil defense at all levels of government. Approximately $13.5 million has been invested by OCDM since 1952 for survey meters, dosimetry and accessory items which have been, or are rapidly being, distributed as basic training and emergency sets to the cities and State civil defense organizations. An additional $2 million procurement program for "standard" radiological instruments was authorized for 1959–60. The program to place operational and training instruments at the local levels of government where they will be most effective in the event of nationwide nuclear disaster is a rapidly expanding one and is backed up with a development program aimed at updating and modernizing the standard items and adding new instruments to fulfill the requirements for a civil radiological defense. In 1959, OCDM was supporting nine development projects for radiological instruments, totaling approximately $200,000. The instrument research program includes projects to develop suitable instruments for general public use as well as for organized civil defense, but procurement of instruments for public use is not contemplated by OCDM.

Dosimetry

OCDM procures dosimeters in three standard ranges for measurement of total integrated exposure to γ-radiation: 0–200 r (CD V-742), 0–20 r (CD V-730) and 0–200 mr (CD V-138) for training and for dose-rate measurement. These are of the self-reading, quartz-fiber, ion-chamber type requiring an external charging unit (CD V-750) for adjusting to zero. Approximately 300,000 of these dosimeters have been delivered and are on grant or loan to the States, and an additional 130,000 are on order. Dosimeters and chargers cost $5–6 each in large quantity procurement.

OCDM has recently endorsed similar dosimeters and chargers for use by the general public. A dosimeter with a range of 0–600 r (CD V-746) and a 0–2 r dosimeter with a special scale for rapid determination of dose rates from 1 to 100 r/hr (CD V-736) have been specified. The companion charging unit (CD V-756) uses a flashlight battery. Bendix Aviation of Cincinnati has made this set of three instruments available to the general public for $20.

OCDM is supporting two development projects to incorporate charging devices in quartz fiber-ion chamber dosimeters as an integral part (CD V-731). An additional research project is underway to provide direct reading of dose rates using quartz fiber in chamber techniques, without requiring an auxillary timing device (CD V-726B). This research and other radiological development projects described in this report were scheduled for completion during the spring and summer of 1960.

Survey meters

OCDM purchases three different survey meters for the measurement of dose rates. The present survey meters cost between $20 and $35 in large-quantity procurement.

The CD V-700 survey meter uses a geiger tube as a detector and has ranges of 0–0.5, 0–5 and 0–50 mr/hr. The probe assembly has a β-shield which may be closed for measurement of γ-radiation only.

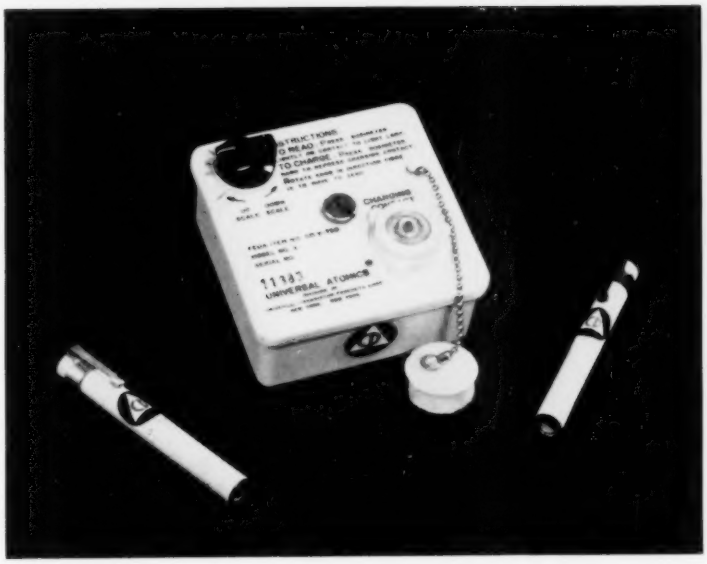


FIG. 1. Dosimeters and charger.

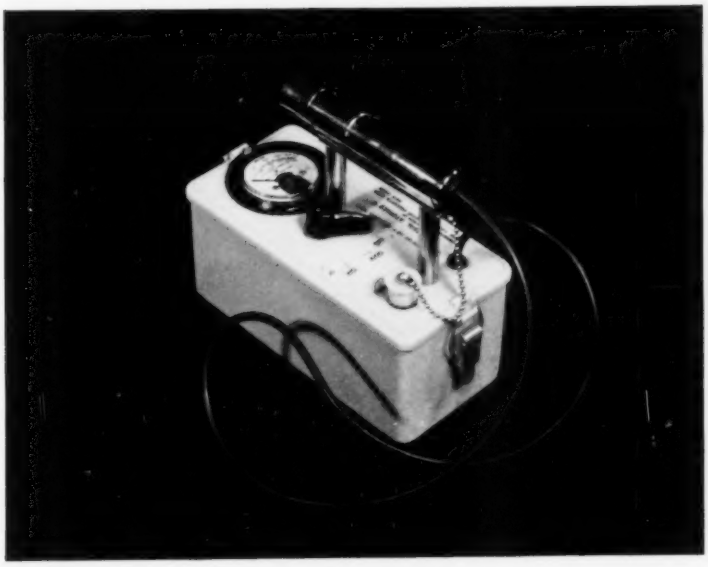


FIG. 2. A CD V-700 survey meter.

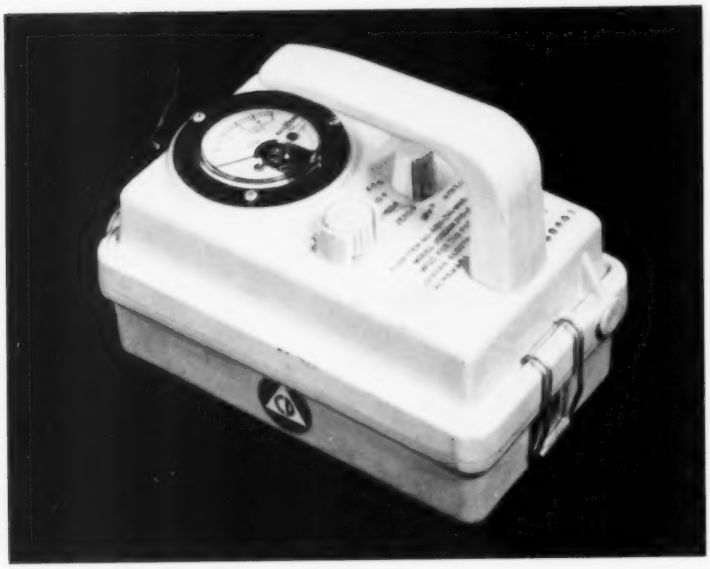


FIG. 3. A CD V-710.

FIG. 4. A CD V-720.

FIG. 5. A CD V-457 demonstration unit.

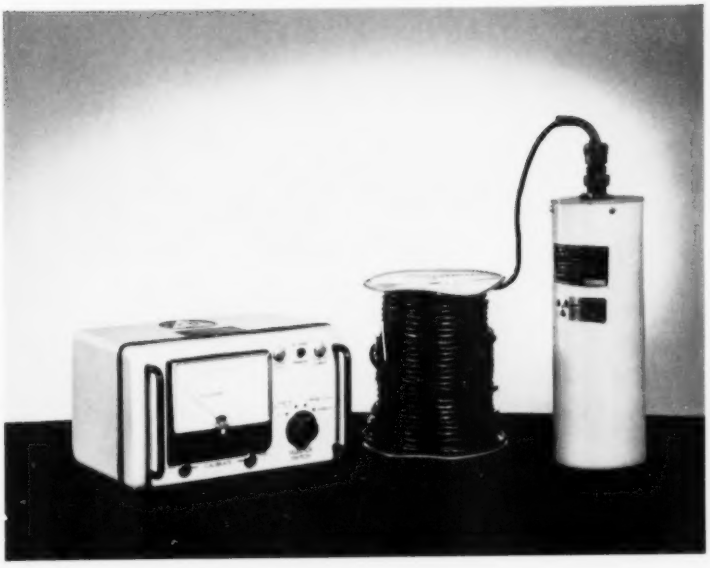


FIG. 6. A CD V-711.

FIG. 7. A complete training kit.

This instrument was developed for use in personal decontamination and for checking food and water. It is also used for training purposes. Over 55,000 of these instruments have been delivered and 30,000 are on order.

The CD V-710 is an ion-chamber instrument for measurement of γ-radiation only. It has ranges from 0–0.5, 0–5 and 0–50 r/hr. Over 170,000 of these instruments have been delivered. This instrument has been termed the "work horse" of civil defense, in that it fulfills the need for a reliable, inexpensive general purpose instrument for local surveying requirements of organized radiological defense teams. Future models of this instrument will have an additional range of 0–500 r/hr and will be designated CD V-715.

The CD V-720 survey meter also uses an ion-chamber detector. It has ranges 0–5, 0–50 and 0–500 r/hr. This instrument may be used to measure γ-radiation, or by opening a β-shield can be used to determine the contribution of β-radiation. About 45,000 of these instruments have been delivered. OCDM is supporting a development project (CD V-716) to incorporate an additional range, 0–0.5 r/hr.

Another research project (CD V-726A) is underway to develop a survey meter for possible public use. This instrument will have a Geiger tube, as the detector and will measure dose rates to 100 r/hr. It will feature high reliability and low cost.

Accessory items—special development

OCDM purchases several additional standard items which are primarily required for radiological defense training programs.

The CD V-457 demonstration unit is a classroom instrument using a Geiger tube as the detector and operating from 110 V a.c. It is used to demonstrate radiation phenomena and to train in measurement techniques. About 300 of these instruments have been purchased, and 1300 are on back order.

About 1000 source sets comprising 30 mc of cobalt-60 and lead shipping or storage containers have been purchased.

An example of an instrument, which is in the second phase of development to eliminate certain deficiencies prior to general distribution through the civil defense system, is the remote-reading shelter meter, (CD V-711). The detector is a pressurized ion chamber meant for installation outside a shelter. A 200-ft cable connects to an inside control unit. It has two ranges each of three decades: 1–1000 mr/hr and 1–1000 r/hr. The CD V-711 is expected to find application in large public shelters, and control and operational centers.

The CD V-780 aerial survey meter is also in the continuing development category. Prototype instruments have been developed and tested. The Atomic Energy Commission, through the Oak Ridge National Laboratory, has recently established a project to further improve this instrument. Aerial survey meters are required at all levels of organized civil defense to enable rapid and safe reconnaissance measurement of radiation levels over large areas, by utilizing locally organized aircraft patrol teams, such as the Civil Air Patrol. It is expected this instrument will enable measurement of ground-level dose rates up to 500 r/hr, at altitudes to 1000 ft.

OCDM is supporting the development of a calibration unit (CD V-794) which could be safely used at all levels of civil defense to maintain accurate calibration of all the standard operational radiological instruments. The calibrator will utilize a 90-c cesium source, will be transportable and will feature interlocking access doors and shields for optimum safety.

Future plans

This brief outline of the OCDM activity in the radiological instrument procurement and development field indicates that our charter is rather broad; we expect to proceed to the development of more sophisticated instruments in the future, as research funds become available. For example, an instrument is required to give quickly a qualitative and quantitative analysis of drinking water. The identification of harmful isotopes is of particular concern to cities receiving their drinking water from open reservoir systems.

OCDM expects, in the next 5–7 years, to procure some $2\frac{1}{2}$ million survey instruments, a like number of dosimeters, and, in addition, training and accessory items for organized radiological defense. The extent and speed with which this program may be implemented depend upon the funds which are made available.

It is likely that an ever increasing number of personal instruments, survey meters and dosimeters will be purchased by the general public in the next few years.

Therefore it is important that, through training programs and information from competent sources, the organized radiological monitors and the general public thoroughly understand the hazard and defense against fallout. It is here the health physicist can make important contributions.

ROBERT B. MARTIN

Instrument Development
Office of Civil and Defense Mobilization, Battle Creek
Michigan

Equipment for Self-monitoring in a Plutonium Facility

(*Received* 11 *April* 1960)

EARLY detection of contamination is one of the primary goals of a Health Physics group in a plutonium facility. Unchecked contamination can lead to unnecessary personnel exposures by ingestion or inhalation. Plutonium may be transferred from hands to food or from surfaces into the air. The control of contamination is also a matter of economics; for example, a defective dry-box glove can lead to the contamination of additional gloves and other equipment if not detected early, thereby causing an unnecessary loss in man-hours and supplies involved in decontamination operations.

While the responsibility for detecting personnel contamination falls primarily on the Health Physics group and its representatives in the work area, the degree of coverage by Health Physics personnel can range from one extreme of complete reliance on Health Physics for contamination detection to the opposite extreme of complete self-reliance by the operating personnel. A justifiable solution lies in compromise, that is, a program of self-monitoring concurrent with continual coverage by trained Health Physics personnel. It is the operator's responsibility to monitor his hands frequently and his clothing occasionally and to notify the Health Physics monitor when contamination is detected. The monitor's responsibility is to maintain continuous surveillance of the work area, to make a comprehensive survey of the operator at the close of a work period or when contamination is detected and to provide contamination information during decontamination operations.

The benefits gained from this compromise plan of providing self-monitoring instruments in addition to Health Physics personnel are many. To the budget minded, it is a much more economical means of providing complete coverage than by providing Health Physics personnel in the numbers necessary to afford absolute coverage. Supplemental instrumentation for self-monitoring frees trained personnel for detailed surveys and assistance when contamination is detected. Providing sufficient self-monitoring instruments assures that there is instant coverage when and where the operator is processing plutonium.

The effort to provide adequate self-monitoring equipment at Rocky Flats has resulted in three instruments, each suited for a distinct application. These instruments are: (1) a modification of the Eberline Instrument Corporation's PAC-1A; (2) Combo, a battery-powered console-type instrument; and (3) a modification of the NRD Instrument Company's CRM-560 Count Rate Meter.

As all of the work with plutonium is done in dry boxes, a compact, lightweight instrument is needed for the dry-box worker. This instrument should be portable enough to be easily moved from one location to another. Fig. 1 illustrates the PAC-1A. as modified for this application. The instrument has a rigidly mounted air proportional probe with the sensitive side exposed. Mounting brackets are placed at strategic locations in the working area (Fig. 2). The lightweight, battery-operated PAC-1A is fitted with a hanger for easy attachment to these brackets. The operator simply hangs the instrument on the bracket nearest his work. The operator checks his hands whenever he leaves the dry-box gloves.

The PAC-1A has a phone output, but for this application only the meter is used. The time constant on the most sensitive scale is short enough to provide an easily seen deflection for each count.

The instrument is light enough to move over coveralls and other surfaces, but the application is awkward owing to the rigidly mounted probe.

Frequent use of the PAC-1A provides early detection of contamination caused by defective dry-box gloves. These instruments are provided on the basis of approximately one instrument per production operator. An estimated modification cost of ten dollars per instrument includes several extra brackets.

To supplement the PAC-1A, a more versatile instrument named Combo was developed (Fig. 3). The Combo is a mobile, battery-powered instrument which utilizes two air-proportional probes, one for use in monitoring hands and clothing, the other for monitoring shoe covers.

The electronic components of the Combo are housed in a waist-high cabinet with the operating controls and readout devices displayed on a slope-front enclosure at the top of the cabinet. In addition, the cabinet provides storage facilities for Health Physics equipment in the process area. An oversize cabinet was chosen for Combo not only to provide storage, but also because of its conspicuous and purposeful appearance. The instrument itself is a reminder to the operator to stop and check himself.

The basic electronics of the Combo were salvaged from the Pee Wee (Nuclear-Chicago, Model 2111) with an additional circuit providing readout from a speaker and a light as well as a count-rate meter. The light, flashing with each count, is useful in noisy areas. Separate amplifiers are used for each detector with a spring-loaded toggle switch provided on the face of the enclosure to select the detector to be used. The switch must be held depressed to activate the detector used for shoe-cover monitoring. The hand

FIG. 1. α-Counter PAC-1A.

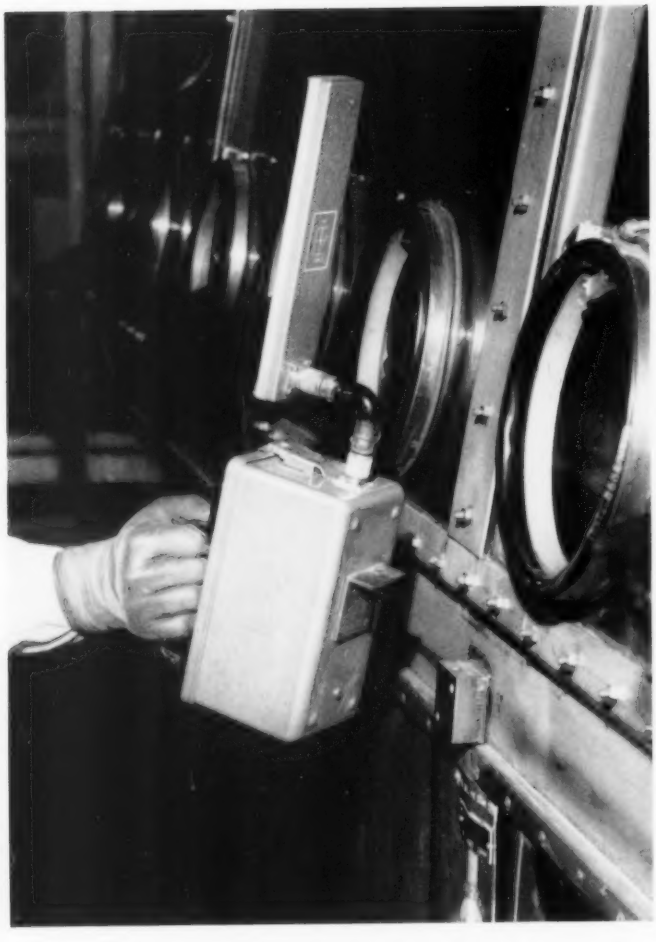


FIG. 2. Mounting brackets for PAC-1A.

Fig. 3. Combo.

Fig. 4. Hand monitor CRM-560.

and foot probes are on separate circuits primarily to avoid adding foot-probe background to hand monitoring.

The probe used for monitoring hands, a commercially available air-proportional detector, is placed face up in a bracket on the operating panel of the Combo. The probe is attached to the instrument by a four foot cable and can be used for clothing survey.

A 6 × 14-inch air proportional detector is used for monitoring the soles of shoe covers. It is mounted in a recess in the cabinet base at a slight angle for user convenience, and to allow contaminated particles to slide off the sensitive area. A guard consisting of three $\frac{1}{4}$-in. rods is mounted to protect the Mylar window. The guard is lifted off to permit wiping dust off the window.

A detachable α-source of approximately 1500 counts/min is mounted in a clip on the operating panel of the instrument for field testing.

The Combo has proved to be especially valuable during contamination incidents. Carrying emergency equipment, several units are wheeled to the perimeter of the contaminated area to relieve the Health Physics monitors of much of the routine personnel monitoring.

The Combos are provided on the basis of approximately one instrument per ten process operators.

The cost of parts and labor for construction was approximately $400 per Combo. This figure does not include engineering or the cost of the Pee Wee.

The final phase of the self-monitoring program occurs when the workers monitor their hands at the locker room exit after washing. A row of modified a.c. powered count-rate meters is provided for this purpose (Fig. 4). CRM-560 count-rate meters have been modified to provide sufficient sensitivity and high voltage to use two 6 × 14-in. air-proportional detectors. The detectors are mounted in angled brackets on top of the instrument housing. The user hears a loud pop and sees a meter swing upscale for each count detected.

In routine usage, all three of the instruments are capable of showing the absence or presence of gross contamination. Spots of α-contamination as low as 400 counts/min (at 2π geometry) are commonly detected. The minimum detectable amount is not defined, since the major parameter is the time that the user spends in surveying a given area.

The instruments are checked daily by an electronics technician. The two most common causes of failure are expended batteries and accidentally contaminated probes. Both the PAC-1A and the Combo routinely run for 1000 hr of operation on a set of batteries.

A controlled atmosphere in the process area permits the use of an air-proportional probe as a detector. The detector has a 0·25-mil entrance window of Mylar coated on both sides with aluminum. The Mylar is stretched smoothly over the sensitive side of the 6 × 14-in. probe to provide a surface which can easily be decontaminated by wiping with a damp cloth.

Personnel unfamiliar with nuclear instrumentation find this equipment easy to use. Simplicity of controls and readout devices contribute to the acceptance of these instruments.

The implementation of a self-monitoring program in conjunction with Health Physics monitoring personnel has proved to be quite successful. Proof of this is evident in the many cases where contamination has been detected by the production worker before spreading has occurred. Indeed, widespread contamination due to routine operations is now an unusual occurrence.

Acknowledgment—Mr. Carl Nordin contributed many valuable suggestions and was responsible for much of the mechanical work involved in this work.

R. D. Smith*
V. P. Johnson

Health Physics Department
The Dow Chemical Company
Denver, Colorado

The Role of the Health Physicist in a University

(*Received* 11 *August* 1960)

Ionizing radiation is perhaps the most recent major factor in the environment which warrants consideration as a public health problem. Health physics, the science of protecting people from the damaging effects of such radiation, has "come of age" as an environmental health activity and must be reckoned with as one which has important public health significance.

At the University of Minnesota the protection of students and staff from the hazards of the physical environment is the concern of the Division of Environmental Health and Safety of the University Health Service. The University Health Service is a separate administrative unit and is responsible to the academic Vice President. The Health Service is closely associated with the Medical School and the medical staff of the University Hospitals.

* Presently with Coors Porcelain Company, Golden Colorado.

The program of the Health Service may be divided into two areas:

(1) The care of illness and the prevention of disease in students.

(2) The supervision of the factors of the physical environment on campus which could be detrimental to the health and well-being of students and staff.[1]

The Health Service is unique in that it has been designated as an official local health department by the Minnesota State Board of Health. Consequently, it has public health responsibilities for some 35,000 students and staff who make up the University community. The environmental health and safety problems of this community are out of proportion to its size because of the nature and diversity of its programs.

Though the hazards of the physical environment have long been of concern to the University, it was not until 1949 that the Division of Environmental Sanitation was established as a part of the Health Service. Its original concern was in the field of food sanitation, student housing and swimming pool sanitation. These services initially were provided by a public health engineer. In 1953, an industrial health engineer was added to the staff and soon thereafter a health physicist. With the recognition of accidental injuries as a public health problem, a safety engineer was also added to the growing staff of specialists.[2]

Today the Division of Environmental Health and Safety, as it is now called, includes, as well as those mentioned above, two additional health physicists, five sanitarians and a bacteriologist.

There is a close working relationship among the professional members of the Health Service staff. The physicians have grown to rely on the staff of the Division of Environmental Health and Safety for follow-through on clinical cases where environmental factors may have contributed to disease or illness. For example, the industrial health engineer may be called upon to substantiate the physician's belief that dermatitis of the hands was the result of exposure to a solvent used in a campus laboratory.[3] If a leukopenia is demonstrated in a physical examination of a linear accelerator employee, the health physicist will be asked for information relative to his radiation exposure history. The medical staff and environmental health personnel mutually benefit by the exchange of information in the evaluation of a public health problem.[4]

Health physics as a function of the University Health Service implies service to rather than a police type surveillance over users of radiation emitters. Serving in an advisory capacity to the Health Service health physics program is the University Advisory Radiation Hazard Control Committee. This committee is appointed by the President and is composed of the Vice President, Superintendent of the Physical Plant and the Director of Radiation Therapy as well as a number of University professors. This Committee is charged with insuring that there is compliance with regulations of the AEC, the State Board of Health, and the University health physics program. Functioning as a service organization, however, the health physics philosophy has been to offer service, advice and assistance to all radiation project directors; this approach has been found successful in creating a general feeling that the health physicist is one who "assists" rather than one who "enforces" and "regulates". The health physicist is someone who is called on for assistance in providing a safe project rather than one who is avoided for fear of revealing poor laboratory techniques or inadequate facilities. It is important that the health physicists establish rapport with all who work on or direct projects involving the use of radiation emitters. It is also important that a good working relationship be established with many departments of the University including the Purchasing Department, the Physical Plant and the research co-ordinator.[5]

At the University of Minnesota, the health physicists assist prospective isotope users with the completion of their application for a by-product material license. This may be the first contact between the project director and the health physicists. It is at this time that a review will be made of the laboratory facilities in which the radioactive materials are to be used. As a result of requests by a number of department heads for assistance in the design of appropriate isotope laboratories, the health physicists have made this service a basic part of the program. It is desirable, when new buildings are constructed, that consideration be given to the establishment of a central isotope laboratory to be used by all who do radiation research within that department. A number of such laboratories have been designed by the health physicists with the co-operation of other specialists in the Division of Environmental Health and Safety. A complete laboratory can be designed by this health team which will insure consideration of all factors of the environment as they relate to safe laboratory operation.

The health physicist is concerned with the safe use, storage and disposal of the radioactive materials to prevent personal injury and area contamination. The industrial health engineer, however, will evaluate the plan from the standpoint of proper local and general ventilation as well as industrial health aspects of laboratory operation. The public health engineer, through his knowledge of water supply and waste handling, provides the information needed to determine

dilution factors likely to be available for the dilution of short half-life radioisotopes in the sewage stream. The safety engineer, another member of the team, might provide specifications for monorail cranes for the movement of lead shielding containers, and suggest procedures for fire safety and safe handling of toxic solvent materials. These specialists work together toward a complete design giving consideration to all aspects of the environment eliminating delays required by lengthy memoranda and other interdepartmental communication.

When the licensee has received his license approval, the health physicist is notified of this approval by the State Board of Health. When an order for radioactive material is placed, the licensee forwards the purchase requisitions to the health physicist for approval. The requisition states that the isotope will be delivered to the Health Service where it is logged in, surveyed, and checked for condition on arrival; it is then delivered promptly to the licensee.

By the time the licensee is ready to use the radioisotope in his research project, he will have at least four contacts with the health physicist. If his project involves the use of hazardous materials in addition to radiation emitters, he will also be contacted by other members of the environmental health team.

Each specialist, being informed on the important aspects of environmental health through his close association with the activities of other members, can be instrumental in bringing about surveys or consultations by other members of the environmental health team. It is this close working relationship that insures more complete environmental health service than could be possible through unco-ordinated individual efforts. The health physicist who operates out of the physics or chemistry department, for example, cannot experience the advantages afforded a group interested in the total environment. The Division of Environmental Health & Safety has recently been allowed to review all requests for University research grants. This affords each member an insight into proposed projects which may present special health problems related to their field.[6]

Another integral part of the University health physics program is personnel monitoring. Personnel monitoring devices are provided to assist the health physicist in evaluating external exposure hazards. Exposure records are kept and forwarded to department heads or project directors with a letter of interpretation by the health physicist.

Radioactive waste, other than small quantities of short half-lived isotopes, are collected in approved, well marked, metal containers. These containers have removable plastic liners allowing the health physicist to collect all such waste with no contamination hazard to the laboratory occupants or to himself. Records are kept of all radioactive waste, whether the waste be contained in paper, research animals, or in toxic solvent materials. These records are kept in compliance with the Health Service's AEC Waste Disposal License, which transfers the waste isotope from the user to the Health Service.

Approximately seventy laboratories on the University campus use a wide variety of radioisotopes for research. These are not used in large quantities; however, the variety of projects and the diversity of activities present interesting and challenging problems for the health physicists. In most of these laboratories, the danger from excessive external exposure does not exist—the danger lies in the possibility of spread of contamination within the laboratory or even within a specific department.

X-ray machines used for therapy, diagnosis and research are inspected and surveyed routinely by a health physicist to insure safety for the physician, the technician and the patient. Plans for X-ray installations are reviewed to provide safe initial installation and to minimize subsequent problems.

Handling spills and other emergencies involving the loss or release of radioisotopes is another important part of the health physicist's work on the University campus. Wrecking tools, air line respirators, self-sustaining air packs, and a wide variety of radiation survey instruments are available for all campus radiation projects through the services of the radiation protection program. The complete removal of contaminated laboratory equipment including walls and floors has been accomplished by the health physics staff of the Division of Environmental Health and Safety.

The following is one example of an emergency that the health physicist was asked to handle recently. Inadvertently, a shipment of radiophosphorus tagged Di-isopropyl Flurophosphate was shipped directly to a licensee. When the shipment was opened, the user noted that a vial containing 100 ml of the chemical had broken inside its glass container. This chemical is a nerve gas so lethal that only very small quantities of it in contact with the skin will cause death. The health physicist received a prompt call from the project director describing the incident. He and the industrial health engineer proceeded to the laboratory where they inactivated the material by proper chemical neutralization, and removed the radioactive remains for proper disposal. The Interstate Commerce Commission was notified of the extreme danger of shipping lethal material in improperly labeled and completely inadequate containers. The central receiving and pre-delivery check of shipments

by the health physicist precludes such occurrences in most instances.

The radiological health services do not end with the safe delivery of the radioisotope to the licensee. Radiation caution signs are displayed on laboratory doors and on storage containers, lab equipment, and on any laboratory device which may present a radiation hazard to personnel. Laboratories are monitored periodically to insure compliance with the Atomic Energy Commission and University operating procedures. Air samples are taken in laboratories where there is any possibility of airborne contamination. Air samples are also taken during the removal of ashes from the radioactive waste incinerators and in the central Hospital isotope laboratory where sources are prepared for radiation therapy.

The increased use of radiation emitters in the field of medical therapy and diagnosis makes the University Hospitals of special interest and concern to the health physicist. The health physicist is notified of all patients who are to receive therapeutic doses of radioisotopes. He is present during the administration of the material and makes recommendations, on the spot, to insure safety for Hospital personnel.

The College of Medical Sciences has established an isotope committee which serves to review all isotope requests by persons in this college who anticipate the use of these materials for diagnosis, research or therapy. This committee was formed at the request of the AEC to comply with the requirements for such a group under the application for a broad medical science by-product material license.

The Director of the Radiation Therapy Department, who is chairman of this committee, directs a memorandum to prospective users answering their request and indicating the need for compliance with the existing health physics procedures prior to submitting their isotope order. The health physicist is also a member of this isotope committee and has outlined the requirements for the users to protect the Hospital environment and the well-being of the medical science staff.

Isolation of special contagion patients who are on radioisotope therapy poses special problems not only for the health physicist but also for the bacteriologist, the newest member of the environmental health team. Because the bacterial infection may present more serious problems than the radiation isolation, it is important that the health physicist recognize this and be informed of the need for possible modifications of his own requirements.[7]

The use of radium sources for therapy presents special problems with regard to their storage, preparation and transportation. A 25 mg radium capsule was lost from the obstetrics and gynecology ward and necessitated immediate surveys of a number of Hospital areas including the laundry, incinerators and waste collection areas. This was accomplished simultaneously by members of the environmental health team. These surveys indicated that the source was not in these locations, and consequently presented no hazard to Hospital employees. The familiarity of the public health engineer with the sewerage system was of assistance in locating the source in a stilling basin in a drop manhole 80 ft under the Hospital. A sewage surge dislodged the source from the stilling basin and resulted in its travel through the sewerage system. Periodic surveys were carried on at the sewage treatment plant, and ten months later the health physicist was successful in reclaiming the source at the treatment plant fifteen miles from the University. It was through the joint efforts of the health physicist, public health engineer, and the industrial health engineer that the radium was eventually found and returned to the University Hospitals.[8]

With reference to joint efforts, mention should be made of the importance of the day-to-day contacts that are made by all members of the environmental health staff. A contact by the safety engineer anywhere on the campus may open the door for the health physicist;[9] a contact by the health physicist in the Physics or Chemistry Departments, in turn, may be instrumental in introducing the services of the industrial health engineer. When a laboratory is surveyed, the health physicist might notice the lack of a fire extinguisher or the need for a deluge shower; this information would be passed on to the safety engineer. The health physicist provides information to the public health engineer on methods used to destroy cultures of organisms which could be hazardous to laboratory workers as well as to students. Because of his training in public health and his knowledge of environmental health and safety services, each member of the team can make valuable contributions to the programs of individual team members and on occasion carry on another's activities.

Education is an important part of the daily activities of the health physicist. Every opportunity is taken to instruct isotope users on sound radiological health practices. Periodic seminars are held for physicians and other Hospital personnel on the important aspects of radiation protection. Nurses who move to new positions have established radiation safety procedures at other hospitals based on their training and experience at the University Hospitals.

Every member of the Division is a member of the faculty of the School of Public Health, and consequently has teaching responsibilities. Special courses are taught in Radiation Hazard Control.

Lectures are also given as a part of a number of graduate courses provided to engineers, nurses, medical students, hospital administrators, public health engineers and chemists as well as to other disciplines in which radiological health may play a role. Though it is not the intent to discuss the philosophy of what and how to teach, including the day-to-day experiences of an operating health physics program in one's teaching enriches instruction and makes it more meaningful for the students.

Techniques, equipment and an understanding of the problem are the tools which the health physicist uses to provide protection for those in this nuclear age who could be exposed to ionizing radiation. However, with the limited number of health physicists, it is important that the most effective use be made of trained personnel in programs to reduce potential hazards associated with ionizing radiation. Because of this shortage, the advantages of co-operation and teamwork in carrying out a complete environmental health and safety program are stressed.

To suggest that health physics requires a whole new set of rules in evaluating the degree of the hazard and in providing a safe operating program for people using radiation emitters, would imply that health physics is an entirely unrelated field, and that previous knowledge in public health would not be applicable. Health physics can profit immeasurably by an understanding and an application of relevant public health practices and procedures which have been gained through many years of experience.

It has been the intention of this paper to point out the advantages of the team approach in environmental health with a concern for the protection of people from all aspects of the environment. The team will certainly be strengthened by the new philosophies, ideas and approaches of the health physicist, and the health physicist will profit by the previously evaluated epidemiological techniques of environmental health. The health physicist should work co-operatively with other environmental health personnel to provide a co-ordinated team effort directed toward the better understanding and more complete control of environmental health hazards.

School of Public Health RALPH O. WOLLAN
and University Health Service
University of Minnesota
Minneapolis, Minnesota

References

1. University of Minnesota, University Health Service, October, (1958). (Bulletin).
2. R. G. BOND, *Amer. J. Publ. Hth.* **45,** No. 7 (1955).
3. G. S. MICHAELSEN, *Hospitals* **31,** No. 22, p. 72 (1957).
4. B. R. REITER and R. G. BOND, *Student Med.* **7,** No. 3, 197 (1959).
5. R. O. WOLLAN, *Hospitals* **31,** No. 22, 76 (1957).
6. R. G. BOND, *Hospitals* **31,** No. 22, 66, (1957).
7. L. D. STAUFFER, *Hospitals* **31,** No. 22, 80, 84 (1957).
8. R. O. WOLLAN, *Sewage & Industr. Wastes* **30,** No. 9, 1197 (1958).
9. G. SCHEFFLER, *Student Med.* **7,** No. 3, 207 (1959).

Maintenance and Field Performance of the PAC-G α-Survey Meter under Extreme Environmental Conditions

(*Received* 4 *May* 1961)

ALPHA-monitoring is performed at the Nevada Test Site at altitudes varying from approximately 3000 to 7500 ft above sea level. These altitude changes cause shifts in the α- and β-sensitivity of monitoring equipment. Such shifts in sensitivity can also result from the rapid variations in relative humidity and temperature which occur when an instrument is taken from open-air areas into underground locations. Additional problems are presented by static-electricity accumulation due to low relative humidities, by airborne sand and dust, and by seasonal variations in temperature from 0 to 115°F.

The portable instrument maintenance facility is located at about 4000 ft above sea level. It is at this level that the PAC-G meters are calibrated; however, they may be used in the field at altitudes ranging from about 3000 ft in the Frenchman Flat area to 7700 ft at the top of the Mesa. Experiments at the Nevada Test Site have shown that the curves of Fig. 1 shift about 20 V to the left along the abscissa for each 1000 ft increase in altitude. In addition the voltage spread between the knee of the plateau (A and A^1 in Fig. 1) and the threshold of beta sensitivity (B and B^1 in the figure) is reduced by approximately 12 V per 1000 ft. Here the threshold of β-sensitivity is arbitrarily defined as the probe voltage at which a 9.1 μc Sr^{90}–Y^{90} source in contact with the probe will cause a significant response by the instrument. If these instruments are adjusted, as common practice dictates, so that the high voltage falls in the center of the plateau at 4000 ft, the effective increase in high voltage (100 V) which occurs when the instrument is taken from the calibration facility to the top of the Mesa often drives the instrument response into the β-region. By departing from common practice and

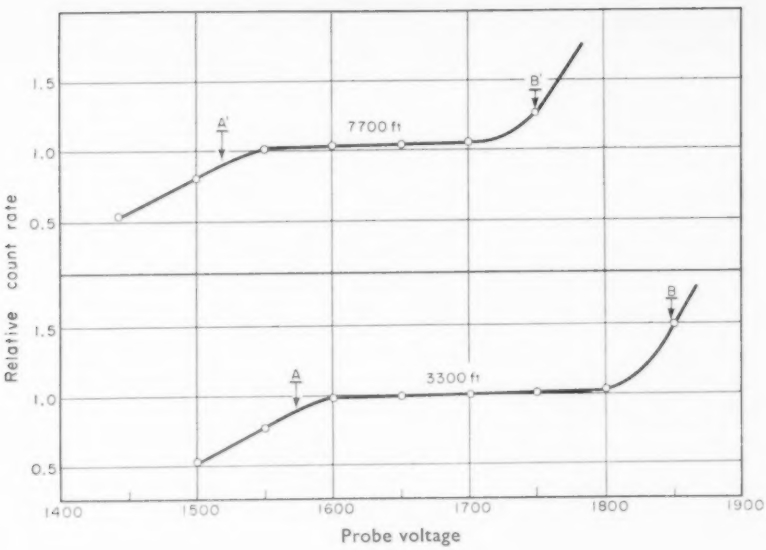


FIG. 1.

adjusting high-voltage supplies to 1550 V, we make the instrument useful over the entire range of altitudes with which we are concerned. It might be well to mention at this point that, although this adjustment places the operating point very near the knee of the curve, high-voltage supplies in these instruments have proved sufficiently stable to prevent a voltage shift large enough to affect the operation.

These instruments must operate in conditions of relative humidity ranging from near zero in the Frenchman Flat area to nearly 100 per cent in the tunnels. In addition they are often subjected on the Flats to winds of gale velocity. The effects of both wind and humidity are minimized by the positive gas pressure within the probe.

Since the probe is not temperature sensitive in the range normally found at the Nevada Test Site, the only problems of significance in this respect are battery life, high-voltage supply and transistor sensitivity. Although the first two of these definitely respond to changes in temperature, neither has been affected noticeably in the two years that the PAC-G has been in use at the Nevada Test Site. Temperature compensating circuits utilized in this instrument have prevented transistor sensitivity changes from affecting normal meter response.

In the early PAC-G instruments, the plastic meter cover became charged with static electricity much too easily. This static charge deflected the meter needle giving a false indication of the meter current. Later models of this instrument, and many of our early models, have been modified by replacing the original meter (a Marion Model MM-3) with a Marion Model HS-3 which is hermetically sealed and has a glass cover. Even in the very low humidity conditions found on the Nevada desert (often below 10 per cent r.h.) there have been no cases of electrostatic charges interfering with readings on the latter meter. The older meters have been made to perform satisfactorily by applying a coat of anti-static spray to both sides of the cover at regular intervals.

One maintenance aid constructed for use with the PAC-G has proved to be particularly useful. It consists of an outer case from a PAC-1A instrument (by the same manufacturer) into which the chassis of any PAC-G may be mounted for trouble shooting or adjustment. Holes have been provided which make it possible to measure high voltage, adjust calibration potentiometers, or plot response curves without the necessity for removing the instrument from the case.

Radiation-Interference Groups H. C. WOODS
BMEWS Project, RCA Service Company
Riverton, New Jersey

Radiological Safety Division F. M. CRANDALL
Reynolds Electrical and Engineering Company, Inc.
Mercury, Nevada

[Note, page 225]

FIG. 1.

Fig. 2.

Tongs Used in Testing for Radioactive Contamination

THE principal hazard of smear-testing for radioactive contamination is that some of the contaminating material may get on the hands of the person making the test. The device illustrated (Figs. 1 and 2) considerably reduces this possibility by making it unnecessary for the hands to come near the area to be tested.

Ordinary laboratory tongs have been modified by attaching a ring and an insert at the end. The surfaces of the ring and insert are angled slightly so the smear paper will not drop through the ring when the paper is clamped between the ring and insert. The outer surface of the ring is angled to prevent its contact with the area to be smeared. Good surface contact between the paper and the area is provided by a felt pad or blotter paper glued to the lower face of the insert, which extends below the ring. Coating the tongs with strippable paint aids in decontamination, if necessary.

T. G. HOBBS

National Bureau of Standards
Department of Commerce
Washington, D.C.

Health Physics Pergamon Press 1961. Vol. 6, pp. 226–227. Printed in Northern Ireland

LETTERS TO THE EDITOR

(*Received* 28 *February* 1961)

EVERY Fall, people engaged in atomic energy work invariably receive requests to assist young students with various school-co-ordinated science fair projects. These requests may be simply for available literature or may be to borrow radioisotopes and outlines of experiments showing their potential uses.

We recently received a request from an advisor to lend a student a radiographic source which he could use to demonstrate "The Principles of Making a Radiograph". He also wanted to borrow some counting equipment to show "Methods of Detecting Radioactivity". After declining both requests it was suggested that a lecture–demonstration using low-level isotopes would be sufficient for the student's needs in the demonstration. One of the students brought some film holders thinking that he could obtain a few radiographs. The detection instruments were demonstrated using calibrated sources of extremely low-level activity. During the course of the demonstration one of the geiger counters was set next to the bag containing the film holders and immediately it indicated the presence of a rather strong radiation emitter. The unknown emitter turned out to be a sealed 56-in length of $\frac{1}{4}$-in Tygon tubing (see Fig. 1), the inter-wall of which was coated with fluorescent radioactive salt (see Fig. 2). When coiled, the tubing emitted 35 mr/hr. The tubing was originally used by the Navy to delineate passageways to blacked-out ships during the war. The boy had infrequently used it as a leash for his cat.

The direct radiation hazard from continuous handling could have been severe, but what was potentially an even greater hazard would have existed if the tubing had ruptured. The salt is in the form of a powder which has a tendency to migrate (see Fig. 2) with handling. A serious inhalation and ingestion problem for the boy and his family would have been present if the cat had clawed through the tubing. This is another instance where materials designed for military purposes can be potential hazards in the hands of civilians.

Figs. 3(a) and (b) are contact autoradiographs of the tubing on type AA Kodak Film exposed for 4 hr and 67 hr.

The balance of the discussion that evening became a question and answer session concerning radiation protection. The science-fair student left to write up his project with a heavy emphasis on radiation protection and without his cat leash.

General Electric Company E. R. HARRIS
Schenectady, New York

Steam-Detergent Decontamination Gun

From many tests and much field experience at Chalk River has emerged a strong belief that high-pressure steam mixed with selected detergents is a powerful decontamination aid. As with most methods, this approach to decontamination does not have universal application, but it has been found to have a very wide range of usefulness. Many objects which are normally wiped or scrubbed clean can be dealt with in less time and, usually, with at least comparable results.

Experience showed that the shape of the nozzle through which the mixture was ejected greatly influenced the measure of success obtainable by the use of this technique. A flat nozzle with a very thin orifice was found to be most effective. Close application of the nozzle to the surface being cleaned gave an effect which can best be compared to a razor blade being passed over face stubble.

The unit is designed to function in the following manner: a high-pressure (at about 90 lb/in^2, gauge) steam supply is drawn from one of the many supply points which are available at the Chalk River plant; some of the steam serves to pressurize a tank which is filled with a detergent solution; the detergent solution is fed slowly into the main steam stream by means of a mixing valve. The mixture passes through

[Letter to Editor, page 226]

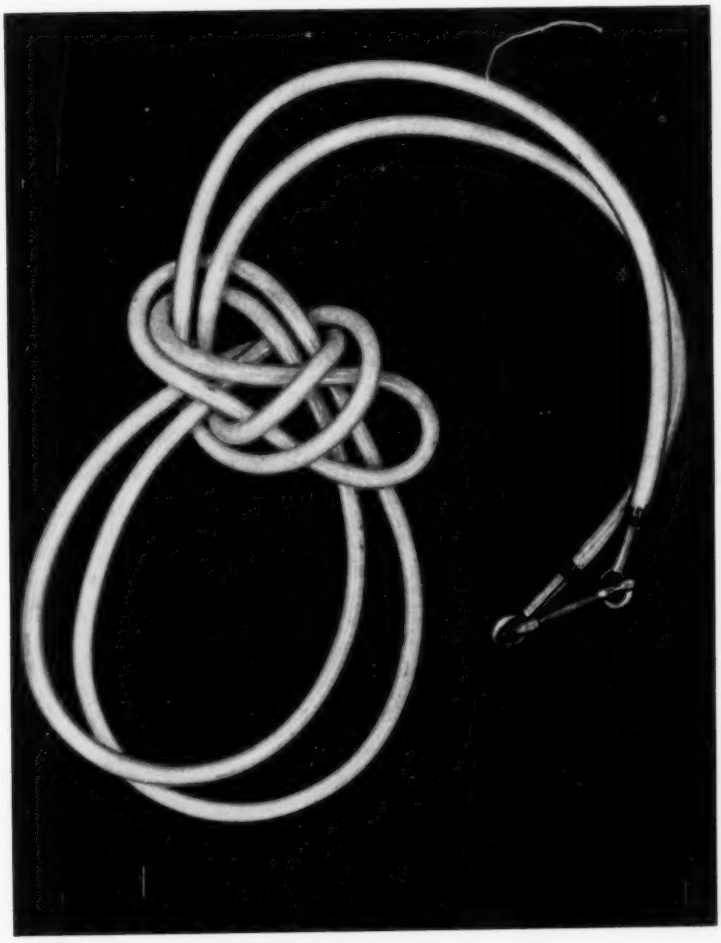


FIG. 1. Daylight photo of guide rope.

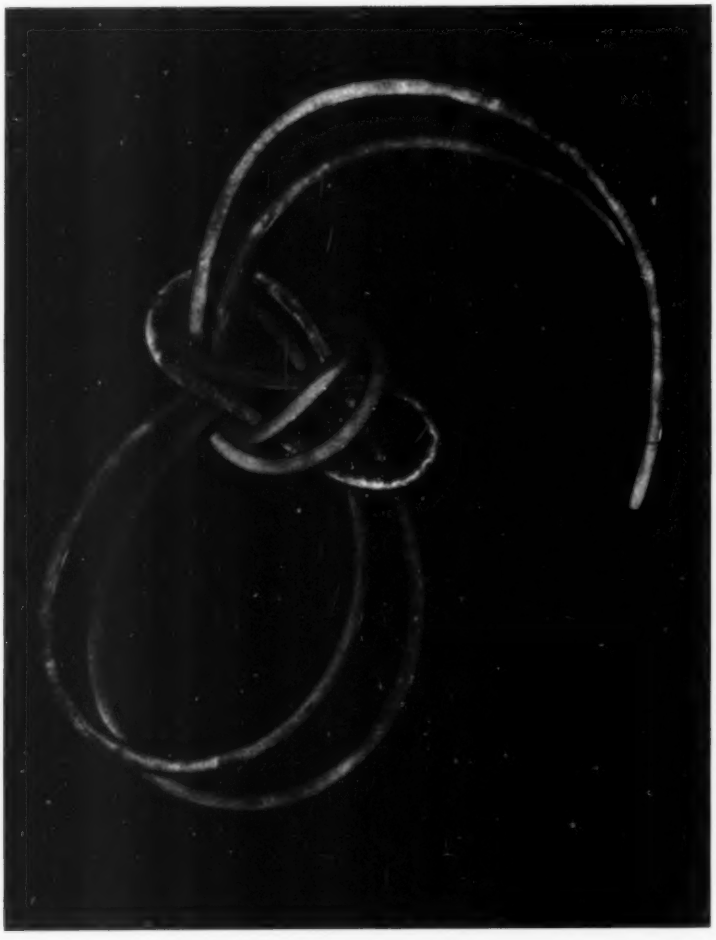


FIG. 2. Autophotograph showing glow characteristics of rope and also showing the migration tendency of the fluorescent powder.

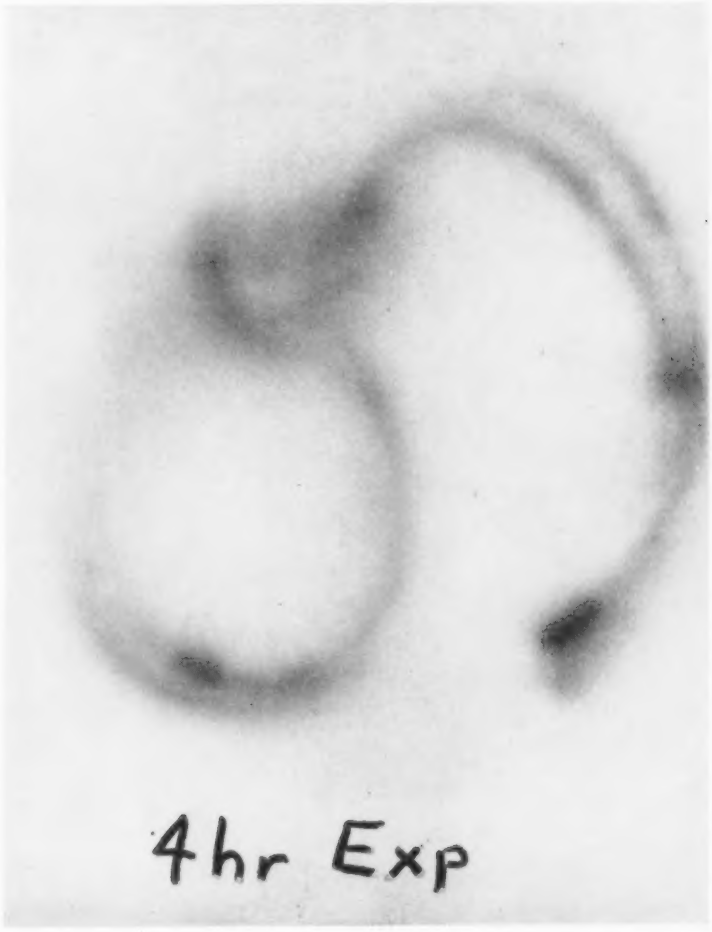


FIG. 3(a).

FIG. 3. Autoradiographs made in flexible film holders using Kodak type AA radiographic film (no lead screens) for (a) 4 hr and (b) 67 hr.

Fig. 3(b).

a light flexible steam hose and discharges through a gun with a flat, narrow orifice. The operation of the unit is controlled by a 6-V electrical system. The detergent flow is approximately 4 gal/hr.

Three types of guns are shown in Fig. 1. All have the characteristic flat orifice with a nozzle opening of 0.015 in. The pistol-type gun is intended for decontamination work carried out at close quarters or in fume hoods; the T-shaped gun with two hand grips is intended for decontamination work on walls and floors; the long gun with the narrow nozzle is intended for use in long-range work.

This equipment has been used successfully on many occasions. One instance was the decontamination of drained fuel-rod storage bays. Another was the cleaning of a series of heavily contaminated small mechanical parts from a reactor loop system.

Chalk River J. NEIL
Ontario, Canada

Health Physics Pergamon Press 1961. Vol. 6, pp. 228–233. Printed in Northern Ireland

NEWS

Editor: J. J. FITZGERALD *Associate Editor:* J. C. COLLINS

Industrial hygienists publish air sampling instrument manual

The ACGIH Air Sampling Instrument Manual is a valuable reference source published by the Air Sampling Instrument Committee of the American Conference of Governmental Industrial Hygienists. It is an up-to-date compendium of instruments for the industrial hygiene field, containing comprehensive descriptive information on the uses, operating principles, physical descriptions, performance data, operating instructions, maintenance instructions and commercial source, if any, for almost all of the air sampling instruments currently in use in the United States. In addition, it contains authoritative discussions on air sampling techniques and theory by distinguished authors including Professor LESLIE SILVERMAN of the Harvard School of Public Health, and THEODORE HATCH of the University of Pittsburgh School of Public Health.

The manual can be purchased through the Secretary Treasurer, ACGIH, 1014 Broadway, Cincinnati 2, Ohio.

American Board of Industrial Hygiene prepares examination for certification

The activities of the ABIH have progressed to the point where examinations are being prepared. A subcommittee under E. C. BARNES, Westinghouse Atomic Power Department, is preparing questions to be given to applicants for certification in the Radiological Aspects of Industrial Hygiene.

Each applicant will be given a "core" examination covering the general aspects of Industrial Hygiene practice in addition to an examination in his specialty.

GE awarded contract to decontaminate and disassemble SL–1 reactor

The AEC has awarded a contract to the General Electric Company, Idaho Test Division, to decontaminate, disassemble and analyze the Stationary Low Power Reactor (SL–1). It has been estimated that it will require seventy men approximately 9 to 12 months to complete the job.

New installation for treatment of radioactive effluents in France

A new installation for the treatment of radioactive effluents will shortly be established at the Nuclear Study Center at Fontenay-aux-Roses, in the suburbs of Paris. The purpose is to treat the waste from the plutonium laboratories which produce between 15 and 25 m^3 of radioactive discharge per day involving 6 c of activity per week. Other smaller quantities of radioactive effluents at Fontenay-aux-Roses and possibly at Saclay, will also be processed.

Essentially, but with certain incidental modifications, the evaporation method developed at Shippingport and other American laboratories will be used.

AEC asks public comment on petition to exempt lock illuminators containing tritium

The Commission has received a petition requesting an amendment to AEC regulations to exempt from Commission licensing requirements automobile lock illuminators containing up to 15 mc of tritium. The tritium would provide luminosity designed to help in inserting a key into the lock at night.

The tritium used in the lock illuminator is in the form of paint completely sealed in plastic. There could be no dispersion of the material into the environment without destroying the plastic ring into which it is incorporated. There is no detectable external radiation from the lock illuminator; thus, an individual lock illuminator containing up to 15 mc of tritium would not present a radiation hazard provided it were manufactured according to specifications which the Commission would establish in a specific license. It does not appear that the distribution of the illuminators would significantly increase natural background radiation exposure to individuals in the population.

The Commission is requesting public comment on the general question of whether it should authorize the use of radioactive materials in consumer goods where control over the disposal of the radioactive materials cannot be exercised, even though the radiation dose to individuals in the population may

FIG. 1. Steam-detergent decontamination guns.

BLANK
PAGE

be extremely low compared with natural background radiation.

If all potential uses for tritium in luminous products were authorized, it is conceivable—but extremely unlikely for the foreseeable future—that the total quantity of tritium which might be distributed annually in consumer goods could eventually approach the amount of tritium now produced annually from natural sources, such as cosmic radiation (estimated at 8×10^6 c/year). Assuming that 8×10^6 c of tritium were released to the general environment annually from various types of luminous products and that such tritium were distributed uniformly in the environment, the distribution would result in an increase of the average radiation dose per individual in the general population of 0.003 mrem/year, which would be equal to the dose from naturally produced tritium and which would be 3/100,000 of the average gonadal radiation dose of 100 mrem from all natural background doses of radiation.

The Commission previously approved the use of tritium on the hands and dials of luminous watches and clocks. However, in that situation tritium will be used instead of radium, which has been used on watches and clocks for many years. Although the use of radium on watches and clocks has generally been accepted as a useful and safe practice, the substitution of tritium results in a reduction in radiation exposure to persons using such timepieces. The Commission did not, therefore, consider the tritium exemption for watches as a precedent for the use of tritium in consumer products which would be exempted from licensing.

The Commission has given notice that it is considering a petition to exempt from licensing and other regulatory controls lock illuminators containing up to 15 mc of tritium.

All persons desiring to submit written comments for consideration in connection with the petition were asked to send them in triplicate to the Secretary, U.S. Atomic Energy Commission, Washington 26, D.C., within 60 days after publication of notice in the Federal Register on April 26, 1961, and were to discuss the following questions:

(1) Should the Commission amend its regulation, 10 CFR Part 30, to exempt from the licensing requirements of Part 30 the distribution, possession and use of lock illuminators each containing up to 15 mc of tritium which are manufactured in accordance with the specifications incorporated in a specific license issued to the manufacturer by the Commission?

(2) If such exemption is granted, should it be limited to automobile lock illuminators or extended to any luminous source containing tritium manufactured in accordance with the specifications incorporated in a specific license issued to the manufacturer by the Commission?

(3) If the Commission exempts lock illuminators containing tritium, or other properly manufactured luminous sources containing tritium, should the Commission limit the total amount of tritium distributed in such forms to approximately 8×10^6 c/year (the amount produced each year by natural causes) or to some other total quantity of tritium in such forms?

USPHS and AEC develop instructional programs in radiation hazards for firemen

Two new instruction programs for helping firemen learn how to cope with radiation hazards in fighting fires have been prepared by the U.S. Department of Health, Education and Welfare and the U.S. Atomic Energy Commission. The AEC Office of Industrial Relations and the Division of Vocational Education, U.S. Office of Education, co-operated in the effort. The courses were designed for state training programs.

The first instructional program, a four-hour orientation unit entitled "Peacetime Radiation Hazards in the Fire Service", has been prepared. It consists of an instructor's guide, a student manual and twelve illustrative charts for use by the instructor. The unit is intended for presentation to large numbers of professional and volunteer firemen as part of their basic training, and to prepare them for more specialized instruction later.

Material for the second program, a fifteen-hour basic course, will be published in mid-summer. It will include an instructor's guide, student's information manual, student work assignments, and twenty-two charts. This program will enable qualified firemen and selected instructors to train others in their own local communities to deal with radiation emergencies.

Fire organizations can obtain information concerning the radiation training programs from their state supervisors of trade and industrial education or other appropriate state agencies responsible for firemanship training.

Material covering the four-hour orientation unit is available through the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C. The student manual is priced at 30 cents, and the instructor's manual at 35 cents. Extra copies of the illustrative charts will be obtainable in limited quantities from the Trade and Industrial Branch, Division of Vocational Education, U.S. Office of Education, Washington, D.C.

National Science Foundation establishes program in radiation biology for secondary school teachers of science and mathematics

Approximately 11,000 secondary school teachers of science and mathematics, grades 7 through 12, will be able to obtain supplemental training under a new National Science Foundation program.

Three institutes in the field of radiation biology will be sponsored jointly by NSF and the AEC. These will be located at Montana State University, Texas Woman's University and the University of Washington.

A typical in-service institute meets once or twice a week for two or more hours during the entire academic year, with some meetings devoted to laboratory or field work. Participants receive allowances for travel and books, and pay no tuition or fees. Sponsoring institutions receive support for direct costs of operation.

First land-burial site for radioactive waste to be operated by industry

The first commercial land-burial site for radioactive waste (low level) is to be operated 100 miles northwest of Las Vegas, Nevada, by Nuclear Engineering Company, Pleasanton, California. The site will supersede temporary AEC service at the National Reactor Testing Station and Oak Ridge National Laboratory. Looking toward start of operations by July 1, Nuclear Engineering is negotiating a burial contract with AEC and a lease with the state of Nevada covering, for 99 years, an 80-acre site within a 46,000-acre desert area near Beatty, Nevada. The company also expects to build a decontamination and repair facility at the site.

City Health Department of N.Y. reports on X-ray equipment survey

HANSON BLATZ, director of the New York City Health Department's Office of Radiation Control, announced that during the course of a two-year survey of medical and dental X-ray equipment in the city, 92 per cent of the medical machines tested—some 3600—were found deficient in operation or equipment. Of the 2700 dental machines tested, 53 per cent were found to be deficient in one or both respects.

Atomic Industrial Forum comments on site criteria

In June, the Forum's Committee on Reactor Safety sent to the AEC additional comments on the proposed reactor site criteria published by the AEC in the February 11 issue of the Federal Register. The comments, in the form of a suggested revision of the criteria, were prepared by a working group named by the Forum Committee and include the criticisms and suggestions produced by the Committee's initial review meeting of March 17.

In a letter to AEC Acting Director of Regulation HAROLD L. PRICE, the Forum's Committee Chairman, W. K. DAVIS, indicated that the members of the working group believe that the adoption by the AEC of site criteria guides can serve a useful and desirable purpose, provided:

"(1) The guides give due recognition to the importance of the engineering design of a proposed reactor as well as to the population density and use characteristics of the site environs and to the physical characteristics of the proposed site; and provided

(2) The guides are sufficiently flexible to accommodate and take advantage of such new information as may be gained from experience of constructing and operating nuclear power reactors."

The Forum Committee did not suggest any change in the AEC's proposed radiation exposure limits which determine radii for the reactor plant exclusion area and the low population zone around the exclusion area. The exclusion area is described as being of such a size that an individual located at any point on its boundary for 2 hr immediately following the estimated release of radioactive materials would not receive a total radiation dose in excess of 25 rems to the whole body or 300 rems to the thyroid.

Differing from the criteria proposed by the AEC in four important ways, the criteria proposed by the Forum Committee would:

(1) Limit the criteria to power reactors rather than to power and testing reactors as proposed by the AEC. The Committee felt that the siting of testing reactors, which are designed for experimental operations, should be evaluated under a different set of criteria.

(2) Assure that example calculations, if included in the appendix to the criteria, would not be considered an integral part of the criteria. The Committee believed that the use of a single-example calculation, as in the AEC proposal, could obscure the intent of the guides and in practice might result in the application of the arbitrary values contained in the example without appropriate regard to the characteristics of a particular reactor or site under evaluation. The Committee redraft, in an attempt to eliminate this possibility, includes four example calculations instead of the one contained in the AEC proposed guides.

(3) Eliminate the population center distance "of at least $1\frac{1}{3}$ times the distance from the reactor to the outer boundary of the low population zone" on the premise that it is without technological basis. The Committee recommended that this concept be

replaced by a man-rem radiation exposure limit expressed as a function of population distribution and density. In its letter to the AEC the Committee suggested that the AEC or Federal Radiation Council undertake to determine how the man-rem concept could be applied.

(4) Focus more attention on the importance of reactor design, on the safety features to be engineered into a reactor and on the interrelationship between design factors and site characteristics, distance and population density.

The Forum Committee's attention was drawn to the impact that the site criteria guides have had and will probably continue to have on power reactor projects in other nations. Although the Committee recognized the primary responsibility of the guides to meet the needs of public safety in the context of site evaluation problems in this country, it also believed that the AEC should be aware of the impact the guides may have in other countries and on U.S. prestige and leadership in power reactor development.

New York State selects site for radioactive waste

The country's first state-owned storage facility for radioactive waste will be on a 3500-acre site in Western New York, approximately 30 miles south of Buffalo.

Governor ROCKEFELLER announced the selection of the site by the State Office of Atomic Development for a nuclear service center. This center will be operated by a private contractor yet to be selected.

OLIVER TOWNSEND, director of the Office of Atomic Development, reported that approximately twenty-four contractors had expressed interest in operating the nuclear center. It has been estimated that the center may ultimately provide employment for 2000 persons depending on the extent to which industry takes advantage of the facilities. Industrial operations at the center may include the reprocessing of nuclear fuels and nuclear research activities.

AEC publishes first report on Alaskan studies for Project CHARIOT

The Atomic Energy Commission has published a first summary report based on more than thirty scientific surveys made during 1959–1960 in northwestern Alaska in connection with the Commission's Project CHARIOT. The technical report is entitled "Bioenvironmental Features of the Ogotoruk Creek Area, Cape Thompson, Alaska."

Project CHARIOT, a part of the Commission's PLOWSHARE program to investigate the feasibility of using nuclear explosives for peaceful purposes, is proposed as an excavation experiment in the Ogotoruk Creek area.

The thirty surveys provide facts about the plant and animal life in the area. This information, when related to the available radiation effects data from the Nevada Test Site and the Pacific Proving Ground, is used to make a preliminary estimate of the possible biological effects of conducting the experiment at the CHARIOT site.

The Committee on Environmental Studies for Project CHARIOT, a committee of scientists which prepared the report, was formed by the Commission's San Francisco Operations Office for the purpose of making the surveys.

The Commission issued a report, prepared by the special scientific committee, which concluded that the experiment could probably be conducted without hazard to plant and animal life in the Arctic region.

The report places the Commission in general opposition to a committee of scientists and laymen in St. Louis known as the Committee for Nuclear Information. The St. Louis group issued its own study of the safety of the Alaska project and came to the conclusion there might be hidden dangers of radioactive contamination of the food chain in Alaska.

Because of the gloomy turn of events in the test-ban negotiations in Geneva, the developing debate may turn into more than an abstract discussion.

If the talks collapse, the Commission wants to get under way with its program, known as Project PLOWSHARE, to demonstrate the peaceful uses of atomic explosives. The proposal to use atomic explosives to dig a harbor at Cape Thompson on the northwest coast of Alaska above the Arctic Circle is one of the key features of this program.

The report was the first comprehensive technical study issued by the Commission on the bio-environmental problems of conducting the Alaska harbor project, called Project CHARIOT. The report was prepared by the Committee on Environmental Studies for Project CHARIOT—a scientific group created by the commission in 1959—and it was based on more than thirty studies made in the last two years of plant and animal life in the remote Arctic region.

The reports of the Commission committee and the St. Louis group were based largely on the same scientific surveys, but they came to contrary conclusions in several important respects.

In assessing the possible radiation effects on plant and animal life, the Commission committee reached this conclusion:

"Considering the predicted distribution and

characteristics of fallout, the morphological abnormalities, pathological effects or population changes as a result of radiation are not expected to occur in the biota (plant and animal life) of the environment, beyond the throw out area."

"It would appear," the committee said, that radiation effects "would be negligible, undetectable, or possibly nonexistent in areas distant from the excavation."

The Commission report did not deal with one of the key points considered in the St. Louis group's study. This was the particular affinity of lichens, which gain their nourishment from dust in the air, to absorb fallout material, such as strontium-90. Since caribou feed on lichens and Eskimos on caribou, the St. Louis group pointed out there was a danger that fallout from the explosion could contaminate the food chain leading from plants to humans.

Public Health Service reports substantial increase in lung cancer among uranium miners

The Public Health Service has reported that a substantial increase in the incidence of lung cancer has been observed in recent months among uranium miners, and that some two-thirds of existing underground mines appear to have levels of exposure beyond recommended working levels (300 $\mu\mu$c of radon and its daughter products per liter of air) currently being used as a general safety guide.

The PHS estimate was given at hearings of the House Appropriations Committee last month, held to consider a request by health authorities in Colorado and New Mexico for federal assistance in financing programs being undertaken in these states to control radiation hazards in uranium mines. Two PHS doctors reported on the results of environmental and medical studies undertaken by the Public Health Service over the past twelve years, with the assistance of the AEC and the State Health Department of Colorado, Utah, Arizona and New Mexico.

The authors reported that while there is wide variation in the distribution and intensity of exposure of uranium miners, calculations based on the environmental studies carried out to date have led to the conclusion that there is a greater potential radiation hazard in underground uranium mines than in uranium mills. They note that there has been considerable progress in improving conditions through better ventilation and other dust control measures, especially in the newer and larger mines, and that co-operation from the uranium mining and milling industry has been "exemplary" in developing the information on which these improvements are based. Nevertheless, the authors added, surveys continue to show that conditions in many mines do not meet the recommended working levels.

The PHS report stated: "While the occurrence of only six deaths from lung cancer in 13,270 man-years of risk may seem too little evidence for serious discussion, nevertheless, the odds are less than one in twenty that five of these would occur by chance among the 907 miners who have worked underground for 3 years or longer. Implication of elevated lung cancer risk is supported by suggestive trends in the prevalence of abnormal sputum samples collected in 1960. Furthermore, since the close of the latest life-table analysis there have been four more confirmed lung cancer deaths and one additional case diagnosed." The sputum findings have caused "grave concern" among the investigators. The authors note that this lead is being "intensively pursued" to determine its significance, both to the individual miners involved and in relations to mining and exposure.

Chief of the PHS Occupational Health Division, Dr. MAGNUSON, observed that "a continuing program of education and promotion of improved environmental control is being accelerated," noting that recommendations for environmental controls could not wait for complete medical findings, in view of the potential hazard involved.

Recommendations for such environmental controls are embodied in the radiation protection standard for mines and mills approved by the American Standards Association last October. This standard is being utilized in connection with a current study of the problem by the Federal Radiation Council.

Radiation Protection Standards for Reactor Operations discussed at Health Physics Society Meeting

A meeting of the American Standards Association N-7.5 on Radiation Protection Standards for Reactor Operations convened in Las Vegas to review the First Revision of the standard in its entirety. Earlier discussions had been restricted to portions of the report. The Second Revision will be circulated for comments before January 1962.

The "Final Report" will hopefully be completed and approved by the committee at the next Health Physics Meeting in Chicago. Upon request copies of the proposal will be released for comments. Copies may be obtained from the News Editor, J. J. FITZGERALD.

ORNL prepares Hanford Fission Products for first radioisotope generator

A shipment of 120,000 c of Cs^{137} and 20,000 c of Sr^{90} was made from the Hanford Works to Oak

Ridge National Laboratory. The Fission Products Pilot Plant (F3P) at ORNL is now operating on a full-time basis to process this material. The Sr^{90} was being prepared for delivery to Martin Company in the titanate form for placement into a 105-w remote weather station. The Martin Company has the contract to install the radioisotope generator at a station in northeastern Canada for the U.S. Weather Bureau.

It is planned that the installation which is the first of its kind for regular service will be made this summer.

New England Chapter of the Health Physics Society holds annual meeting

The second annual meeting of the New England Chapter was held on May 24, 1961, at the Watertown Arsenal of the U.S.A. Ordnance Corps, Watertown, Massachusetts. The meeting featured a symposium on problems in health physics oriented towards areas of particular concern in the New England area. Speakers were Russ Cowing, Executive Secretary of the Health Physics Society: Shields Warren, NCRP; Charles Williams, Liberty Mutual Insurance Company, and James Terrill, U.S. Public Health Service.

A tour was made of the Arsenal uranium melting and alloying facilities and swimming pool reactor facility.

The evening session featured a talk on "The Role of Man In Space" by Frederick Durant of the AVCO Research Center, Wilmington, Massachusetts.

Health Physics in Norway—L. Berteig

Norway and IAEA co-operate in reactor research program. The experimental zero power reactor NORA, sited on the premises of the Atomic Energy Institute, Kjeller, Norway, went critical the first time on June 14, 1961. The responsibility for the reactor project is shared with the International Atomic Energy Agency. The IAEA last year accepted an invitation from the Government of Norway to take part in a wide research program. In part, this arrangement was made because of the design of NORA, which is built with an extremely simple geometry. For example, exceptional care has been taken to make the bottom of the cylindrical vessel absolutely flat, so as to make it possible to solve the theoretical equations relating to the geometry of reactor cores. In addition the proposal and the arrangements were made because of the interest in the preliminary research program suggested by Norway, which include the measurement of integral reactor physics data of cores moderated entirely by light water or heavy water, and studies on mixed heavy and light water moderators.

Norwegian committee studies entry of Savannah into territorial waters. With a possible visit of the *N.S. Savannah* in view, a special committee in Oslo has studied Norwegian Harbors and territorial waters in relation to atomic energy propulsion in shipping. The studies have proved complex and will probably not be completed before 1962.

Use of high-energy radiation for therapeutic purposes strongly encouraged. Recent technical improvements have made high-energy radiation installations more readily available to medical therapy centers. In the light of the more favorable radiation absorption ratio for bone/soft tissue offered by such installations in the treatment of certain malignant diseases, the State Institute of Radiation Hygiene has made a drive for high energy equipment. Radiation centers are strongly encouraged to supplement their radiation facilities.

Massachusetts Nuclear Incidents Committee

The Massachusetts Commission on Atomic Energy has established an informal nuclear co-ordinating committee composed of representatives of the various governmental and non-governmental agencies and individuals within the state who have an interest and/or a responsibility in radiation protection.

The purpose of this committee is to determine how to utilize best the nuclear capabilities within the state and to co-ordinate them so that they can be effectively used in the event of a nuclear incident.

Subcommittees have been established to investigate and make recommendations relative to potential hazards, emergency transportation, indemnification, training, identification and notification, and equipment and servicing.

MIT research reactor amendment

The Atomic Energy Commission proposes to issue to the Massachusetts Institute of Technology an amendment to their Facility License NO R-37. The amendment will authorize MIT to increase the power level of their research reactor, located in Cambridge, Massachusetts, from 1 MW to 2 MW (thermal) and to make certain modifications in the reactor and operating procedures to accommodate operation of the reactor at the increased power level.

Health Physics Pergamon Press 1961. Vol. 6, pp. 234–252. Printed in Northern Ireland

SIXTH ANNUAL MEETING OF THE HEALTH PHYSICS SOCIETY

13 June to 16 June 1961 at Las Vegas, Nevada

Abstracts and titles of papers presented at the meeting

Education and Training in Health Physics. Moderator: W. D. CLAUS, U.S. Atomic Energy Commission, Washington, D.C.

Dedication to Dr. Elda E. Anderson. J. S. LAUGHLIN, President, Health Physics Society.

Since this symposium on education and training in health physics is the first event on our scientific program and is the subject in which Dr. ELDA ANDERSON had made a major contribution, it is particularly appropriate to speak of her loss at this time.

Members of our Society, and the many other friends and colleagues of Dr. ELDA ANDERSON were shocked and saddened at her death on 17 April 1961.

Dr. ELDA ANDERSON, or "Andy" as we knew her, was a former President of this Society and at the time of her death was Chairman of the American Board of Health Physics. She was instrumental in the formation of the Health Physics Society and served initially as its Secretary–Treasurer. Our Society and the entire profession of health physics has lost one of its most dedicated and effective leaders.

Professionally, Dr. ANDERSON had the satisfaction of an active and successful career. After she earned the Ph.D. in physics at the University of Wisconsin she served successively as a college professor of physics, with the Office of Scientific Research and Development, and with the Manhattan Project at Los Alamos. Since 1949 she had been with the Division of Health Physics at Oak Ridge National Laboratory. During this time she developed the training program in health physics at Oak Ridge and initiated a combined M.S. degree program with Vanderbilt University, which became a model for other institutions. She was also effective in the training of foreign Health Physicists and conducted the International Courses in Health Physics in Sweden, Belgium, and in India. She was both a pioneer in health physics and the primary establisher of educational programs in this field.

Personally, Dr. ANDERSON was always cheerful, friendly, helpful, and effective. She was enthusiastic and a very good friend of all who knew her. It is safe to say that more people engaged in the practice of health physics, and more people in this room, knew her personally than any other health physicist. That this is so is a testimony not only to her technical achievements and her educational effectiveness, but also to her sincerely friendly personality.

The greatest tribute that we can render to her aspirations, and in her memory, is in the standards we maintain and the imagination we display in the practice of health physics, and also in the effectiveness of our efforts to make this Society, to which Andy was so devoted, an increasingly useful and vital organization.

Existing training programs. W. D. CLAUS, U.S. Atomic Energy Commission, Washington, D.C.

Needs in government. F. J. WEBER, Division of Radiological Health, Department of Health, Education and Welfare, Washington, D.C.

Needs in industry. E. C. BARNES, Westinghouse Electric Corporation, Pittsburgh, Pa.

Needs in medicine. W. SEAMAN, Columbia Presbyterian Medical Center, New York, N.Y.

Objectives of academic training programs. M. EISENBUD, New York University Medical Center, New York, N.Y.

International aspects of health physics training. M. SUZUKI, International Atomic Energy Agency.

Certification standards. F. P. COWAN, Brookhaven National Laboratory, Upton, N.Y.

Professional status for the health physicist. W. D. CLAUS, U.S. Atomic Energy Commission, Washington, D.C.

Internal emitters—I. Inhalation Studies. Chairman: J. N. STANNARD, University of Rochester, Rochester, N.Y.

Effect of particle size and total dose on retention and translocation of inhaled plutonium.* W. J. BAIR, Biology Laboratory, Hanford Laboratories, General Electric Company, Richland, Washington.

Beagle dogs were exposed to plutonium dioxide aerosols of three particle size distributions at concentrations from 10^{-7} to 10^{-3} μc per cm^3 of air. For all particle sizes, the percentage of plutonium escaping early rapid clearance and retained for slow clearance increased as the total deposited dose was decreased from 2 mg to 0.02 μg. For a given amount of plutonium deposited, the percentage retained for slow clearance was greatest for plutonium inhaled as particles with a mass median diameter (MMD) of 4.3 μc, less for 3.0 μ and even less for 1.7 μ particles. The subsequent rate of clearance increased inversely as the mass median diameter of the particles. The total amount of plutonium translocated and excreted in urine was greatest for 1.7 μ particles and was apparently related to the particle size distributions. All results favored pulmonary retention of 4.3 μ particles in comparison with 3.0 μ and 1.7 μ particles.

Bronchogenic carcinoma from chronic irradiation with radioactive cerium fluoride ($Ce^{144}F_3$) particulates. H. CEMBER, University of Cincinnati, Cincinnati, Ohio.

Rats were treated by intratracheal injection with 1 μ $Ce^{144}F_3$ particulates at dose levels of 4, 2, 1, and 0.5 μc Ce^{144}, and were permitted to live until death by "natural" causes. Lung cancer was found among rats in all treatment levels. The smallest absorbed dose in the case of bronchogenic carcinoma was 2500 rads.

Despite the fact that the rats were exposed only once, the long pulmonary retention time of the radiocerium resulted in continuous radiological insult to the lungs for the duration of the animals' lives. Pulmonary clearance studies, which were performed in order to compute the absorbed dose to the lungs, showed the deep respiratory tract to behave as a two-compartment model. Clearance from each compartment followed first-order kinetics, and the respective clearance half-times for the compartments were 24 days and 193 days.

Translocation and excretion of inhaled $Ce^{144}O_2$ particles.† R. A. HENNACY (Introduced by WM. J. BAIR), Biology Laboratory, Hanford Laboratories, General Electric Company, Richland, Washington.

Particles of $Ce^{144}O_2$ were administered to rats in the form of an aerosol. $Ce^{144}(NO_3)_3$ was added to carrier $Ce(NO_3)_3$ and mixed with Na_2O_2 to produce sub-micron particles *in vitro*. One week after exposure, about one-third of the deposited dose was in the lung, 3 per cent in the liver and less than 1 per cent in all other tissues. After initial clearance, cerium was slowly excreted in the feces and very little appeared in the urine. Cerium was cleared from the lungs at a very low rate.

Distribution and excretion of thorium in the rat following parenteral administration. R. G. THOMAS, R. LIE and J. K. SCOTT, University of Rochester, Rochester, N.Y.

Past studies on thorium metabolism have indicated a substantial "carrier-effect", with the pattern of distribution and excretion depending both upon the amount of thorium administered and the route of entry to the animal. Such an effect bears directly upon the MPC because of the dependence of biological retention time upon the specific activity of the thorium isotope.

Three solutions of widely different concentrations of added carrier (thorium-232) were made in the citrated form from a thorium-234 chloride solution. Aliquots of these were injected intravenously, intraperitoneally, intramuscularly and intratracheally into rats, and studies were made of the time-course of whole-body and tissue radioactivity, excretion patterns in urine and feces, and kinetics of early blood disappearance.

Results indicate a significant carrier-effect following all routes with the skeletal deposition being highest at the tracer level and the RES localization being highest with added carrier.

Distribution and excretion of thorium in the rat following inhalation. B. B. BOECKER, R. G. THOMAS and J. K. SCOTT, University of Rochester, Rochester, N.Y.

Experiments have been undertaken to study the metabolism of thorium following inhalation.

A small exposure chamber was used which permitted the simultaneous exposure of 20 rats for each experimental run. Various amounts of thorium-232 chloride have been aerosolized with tracer thorium-234 in order to detect alterations in distribution and excretion patterns as a function of carrier. Three rats from each run were counted alive at weekly intervals. Several rats were killed immediately following exposure for lung deposition data and the remainder were placed in metabolism cages for collection of excreta and subsequent serial sacrifice.

Results indicate similar patterns of whole-body retention for the various levels of carrier. Excretion was mainly via the feces although some urinary clearance was observed in the early post-exposure days. A smaller fraction of the body content was found in skeleton with the higher carrier levels.

Inhalation of U_3O_8 smoke from exploding wires. B. R. FISH and F. G. KARIORIS, Oak Ridge National Laboratory, Oak Ridge, Tennessee and Marquette University, Milwaukee, Wisconsin, respectively.

A uranium oxide aerosol consisting of isolated spheres (in the range 0.02–0.1 μ) was prepared with an exploding

* Work performed under Contract No. AT(45-1)-1350 between the Atomic Energy Commission and the General Electric Company, and between the United States Air Force-Delivery Order No. (29-601)-58-3900 and the Atomic Energy Commission.

† Work performed under Contract No. AT(45-1)-1350 between the Atomic Energy Commission and the General Electric Company.

wire aerosol generator and was administered to mice as a single, nose only, inhalation. The animals were sacrificed in groups of 5 at 0, 3½, 7, 14, 28, 56, and 112 days after inhalation. Tissues and excreta were analyzed to determine deposition in body organs and elimination rates. Results are to be compared with data from a similar experiment in which larger particles (about 0.75 μ) of U_3O_8 were used. Preliminary data indicate a much higher deposition in the gastrointestinal tract than was anticipated.

Size-selective samplers for estimating "respirable" dust concentrations. M. LIPPMANN and W. B. HARRIS, U.S.A.E.C., Health and Safety Laboratory, New York.

The HASL has developed and used a family of size-selective air samplers for field survey work. Each operates at a flow rate suitable to a particular suction source used by the HASL, ranging from 0.9 l/min to 40 f^3/min, and consists of a cyclone collector followed by an efficient filter. The collection characteristics of the cyclones are selected so that the fraction of the sample collected on the filter is equivalent to the material which would penetrate to the non-ciliated portions of the lung as defined by the human retention experiments performed by BROWN, HATCH and others.

Representative field data are presented illustrating the relative proportions of "respirable" and "non-respirable" dusts at various operations in the uranium industry.

Maximum permissible exposures. Chairman: WRIGHT H. LANGHAM, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

Variability of relative organ concentration of trace elements in human tissue. W. S. SNYDER and M. J. COOK, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

The first report of the Federal Radiation Council uses the arbitrary assumption that within an appreciably homogeneous population group the majority of individuals do not vary from the average by a factor greater than 3. In an attempt to evaluate this assumption, concentration values of Sr, Al, Cu, and Zn in kidney, liver and lung have been studied. These concentration values were obtained from a spectrographic study of tissues obtained from autopsies of victims of instantaneous death.

To obtain a sample size sufficient to permit analysis at a 95 per cent level of confidence, data on adults from five urban groups were pooled. The ratio of the highest sample value to the sample median has been tabulated, and this sometimes exceeds three. Preliminary statistical analysis at the 95 per cent confidence level indicates that, for most elements and tissues tested, less than 5 per cent of the adult population exceeds three times the median.

Radionuclide classification—Relative and absolute hazard. J. W. VANDERBEEK, Hanford Atomic Products Operation, Richland, Washington.

The logic and limitations to any classification system for radionuclides are discussed. A formula for ranking the relative hazard of radionuclides is presented, relating effective half-life and body-burden values. A means for comparing the absolute hazard between different physical and chemical forms and quantities of radionuclides is also presented.

The estimation of the internal radiation exposure resulting from the ingestion of fallout-contaminated water in a nuclear war. M. B. HAWKINS, University of California, Richmond, California.

The over-all problem in a wartime situation of controlling internal radiation exposure resulting from the ingestion of water contaminated by fallout is discussed. The limitations of maximum permissible concentration concepts in areas where fallout contamination levels are high are discussed and simplified methods of computing the internal dose are presented. To facilitate the interpretation of such information, the "committed dose" concept is introduced and its proposed use is described.

A brief discussion of the significance of the hazards of fallout-contaminated water supplies relative to other hazards in the nuclear war situation is presented.

Comparison of predicted and measured equilibrium levels for chronically administered cesium-137. C. R. RICHMOND, and J. E. FURCHNER, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

The practice of using retention parameters from acute studies to predict equilibrium levels attained during chronic exposure has rarely been subjected to experimental verification. This report provides additional support for such practices.

The retention of an acute oral dose of Cs^{137} by female RF mice was measured by whole-body counting in a 4π liquid scintillation detector and subsequently reduced into a three-component exponential function by a 704 curve-fitting procedure. Increase in activity was also measured in mice which were given 0.012 μc/ml Cs^{137} daily in their drinking water and compared with the predicted build-up. Fall-off of activity following cessation of chronic exposure was also measured. Both build-up and fall-off of activity agreed well with those predicted from parameters obtained from the acute study.

The effect of age on retention parameters and, therefore, on equilibrium levels is a complicating factor which will be discussed.

Development and Design of Laboratory and Plant Equipment. Chairman: P. B. K'BURG; Co-Chairman: H. A. MCCLEAREN, E. I. du Pont de Nemours and Company, Inc., Savannah River Plant.

Radiation design and the health physicist. L. LEWIS, Carolinas-Virginia Nuclear Power Associates, Inc.

A description of the complete misdesign of a Fuel Element Manufacturing Facility, consequent production difficulties and subsequent major corrective measures are outlined in this paper to support the contention that:

(1) The integration of design and operation is vital in all nuclear work.

(2) Operational experience must be supplied to the designer by specialists.

(3) The services of an experienced Health Physicist are invaluable during the design stage to provide necessary operational experience and guidance.

The interrelationships of the design team and the design requirements are discussed briefly to point out the role of the Health Physicist.

Health physics aspects of handling spent fuel elements at the Savannah River Plant. R. D. CALDWELL, E. I. du Pont de Nemours and Company, Inc., Savannah River Plant.

The handling of spent natural and enriched uranium fuel elements from SRP and other sites is discussed, with emphasis on Health Physics problems encountered in the removal of failed fuel elements from the reactor, their storage, disassembly, and inspection, and their preparation for chemical processing. Attention is also given to the routine removal and handling of unfailed elements, and to intra-plant shipping problems.

Health physics considerations in the design of the Argonne 12.5 BeV zero gradient synchrotron. R. V. WHEELER, Argonne National Laboratory, Lemont, Illinois.

The Argonne Zero Gradient Synchrotron capable of accelerating some 5×10^{13} protons per pulse to an energy of 12.5 BeV is due to initiate operation in late 1962. During the planning stages of this facility, Radiation Safety cooperated with the various design groups, incorporating control, warning, and remote monitoring features.

Massive external shielding, 20–50 feet of earth or its equivalent, is required at the Ring Building to reduce the expected stray radiation backgrounds from the synchrotron.

An effective health physics program has been organized to be initiated in late 1961 during initial operation of the linear and preaccelerators.

Radiation and contamination control improvements for a plutonium processing plant. W. L. MARTER, E. I. du Pont de Nemours and Company, Inc., Savannah River Plant.

A new plutonium processing plant was designed, constructed and placed in operation. The design improvements for radiation and contamination control reflect operating experience at the Savannah River Plant as well as other Atomic Energy Commission installations. Improvements incorporated for radiation and contamination control have proved sound in operation.

Safety performance testing for sealed radioisotope sources and devices. P. C. AEBERSOLD and J. W. HITCH, U.S. Atomic Energy Commission, Washington, D.C.

The need for development of techniques as a basis for safety testing of radioisotope devices is well recognized. Current test procedures and fabrication specifications are largely those proposed by individual manufacturers and are not necessarily reliable.

Source safety development and testing is now a part of the AEC Isotopes Development Program. Emphasis is placed upon development of compounds that are biologically inert. Tests are being developed to assure maximum insolubility, heat resistance, and containment of radioisotope compounds consistent with efficient utilization.

Techniques developed will be used as a basis for the establishment of standard testing criteria and source fabrication requirements.

Control of contamination in high level caves at the Savannah River Laboratory. R. C. CRAFT, E. I. du Pont de Nemours and Company, Inc., Savannah River Plant.

Methods of controlling contamination in high level caves at Savannah River Laboratory are presented. A brief summary of recent operating experiences, including cell decontamination procedures, is presented.

Hazards associated with liquid hydrogen bubble chambers. A. A. WEINTRAUB and M. S. WEINSTEIN, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York City.

Along with the benefits achieved through successful use of the liquid hydrogen bubble chamber as a tool in high energy nuclear research, are the hazards attendant to its application. The principal source of danger lies in the malfunction or failure of equipment releasing gas into the atmosphere with the formation of an explosive hydrogen–air mixture. Important safety considerations are proper design of equipment and housing facilities, hydrogen control before, during and after bubble chamber operation, and elimination of ignition sources. Design and operation are complicated by the lack of universal standards for handling liquid hydrogen and the use of conflicting procedures for controlling hydrogen hazards at various facilities.

Experience with an ionization chamber pulse reader at Hanford. F. I. RISING, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

The various applications that have been made of the ionization chamber pulse reader at Hanford are described. The types of measurements made, problems encountered and the results obtained with various ionization chambers are discussed. Some typical applications are the measurement of natural background, air-borne tritium, hand dose, immersion dose in the Columbia River, and *in vivo* dose from internally deposited radionuclides with small chambers.

Radiological development within the Hanford Radiation Protection program. C. M. UNRUH, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

The radiological development activities within the

Hanford Radiation Protection Operation are designed to make prompt use of research results. By separating these application–engineering functions from research activities, field-engineered devices or methods are placed into service at an early date and research efforts receive maximum utilization without undue field detail required from the research staff. Application–engineering functions emphasize the practical application of research results. The current Hanford program of radiological development within the Radiation Protection Operation is briefly outlined to stimulate inter-laboratory exchange of development information.

Calibration of a Fission Gas Monitor. W. C. Day and R. F. Selden, Lewis Research Center, National Aeronautics and Space Administration, Cleveland, Ohio.

A method for calibrating β-sensitive gas monitoring equipment is presented which avoids precise mixing of concentrated active gases with diluent air. The method depends upon neutron activation of the argon in P-10 proportional counting gas and passage of the active gas successively through the monitor chamber and a proportional counter chamber. The specific activity is determined directly by the count rate and volume of the proportional counter chamber. The method is satisfactory for specific activities corresponding approximately to MPC levels.

Radiation safety considerations for subcritical reactors. R. W. Clack and W. R. Kimel, Kansas State University.

From operating experience, a comparison is offered of the contribution to delivered dose rates from γ-, thermal neutron and fast neutron radiation with and without cadmium cover on subcritical assembly. A simple but sensitive air monitor for checking Pu–Be source integrity is described. A method of measuring k_{eff} to assure avoidance of accidental criticality and an anomalous result are presented. Calibration techniques for use with both fast and thermal neutron detectors are described. Relative magnitude of the various hazards associated with subcriticals is assessed.

Miscellaneous Topics in Health Physics. Chairman: M. Eisenbud, New York University Medical Center, Institute of Industrial Medicine.

An experimental evaluation of multiple crystal arrays and single crystal techniques. C. E. Miller, Argonne National Laboratory, Lemont, Illinois.

The γ-ray spectra of humans who contained known quantities of K^{42} were obtained with a NaI(Tl) crystal placed at various positions adjacent to the body. These spectra have been analyzed mainly to determine the spread in counts per minute per microcurie in the body which result when patients of different body builds are measured with the crystal at the various positions. Also, the counting rate obtainable with multiple crystal arrays is compared to that observed with a single crystal. The uncertainties which will result if these arrays are used to measure a human's normal K content, without administering a calibrating tracer quality of K^{42}, are given.

Natural environmental radiation measurements in New York State. W. M. Lowder and A. Shambon, U.S. Atomic Energy Commission, New York City.

During 1960, measurements were made of the penetrating component of outdoor natural radiation in the New York City metropolitan area and forty-one communities in other parts of the State, using portable scintillation detectors and 20 l. air-filled ionization chambers. Most of the readings lie within the range of 9–14 μr/hr observed on earlier surveys as typical of the eastern United States, with indications of trends in particular areas. The data are summarized, with particular reference to the often inferred relationship between natural radiation and geological substructure and to the continuing study of the suggested correlation between natural radiation and the incidence of congenital malformations in New York State.

Fallout strontium-90 in the human environment. A. R. Schulert, Lamont Geological Observatory, Columbia University, Palisades, New York.

The monitoring and assessment of strontium-90 in the human bones from around the world as well as milk, other foodstuffs, and other biological samples from more limited areas have been carried out at the Lamont Laboratory for the past eight years and are being concluded at the close of the present fiscal year, i.e. 1 July, 1961. Bone samples have been obtained regularly from twenty locations and sporadically from fifteen others around the world representing every continent. Twelve thousand have been collected and analyzed, most as single bone specimens, although the majority of the adult bones in the last few years have been combined with others of similar time and locations. In addition, about three thousand milk samples from systematically selected sites have been processed together with several hundred other food samples.

These data now give an adequate assessment of the contamination from bomb tests to date and thus, a basis for prediction for future atmospheric contamination from tests, accidents, or other causes. Since there has been no large scale testing since the fall of 1958, it is now found that, contrary to earlier predictions, the strontium-90 concentration in the diet, for the western culture at least, is significantly reduced from the peak values of 1959. However, adult bones, which are still far from equilibrium with their diet, will continue to rise in concentration. The more critical children's bones, which reach concentrations several times that in the adults, have leveled off and are on the decline.

Preliminary evidence indicates that this decline may be more rapid for the individual on a milk diet (i.e. western culture) than it is for the non-milk diet (i.e. Asian culture).

Sampling networks for radioactivity in air. R. H. Goeke, J. G. Bailey and A. P. Zizos, Division of Radiological Health, Public Health Service.

The Federal Government is conducting many radiation

sampling programs to measure the levels of radioactivity in our environment. Measurements of radioactivity in air are of importance since such programs act as "alert" systems. The discussion will be concerned with those operated by the Public Health Service, Atomic Energy Commission, Air Force, Naval Research Laboratory and the Weather Bureau.

A discussion of each network will include such topics as locations of air sampled, sampling procedures, limits of overall accuracy, instrumentation, and methods for reporting results.

Correlation of uranium α surface contamination, air-borne concentrations and urinary excretion rates. N. B. SCHULTZ, B. F. SHINN and A. F. BECHER, Union Carbide Nuclear Company, Oak Ridge, Tennessee.

In this report are presented statistical evaluations of such field data as work surface α-contamination levels, air-borne uranium concentrations and the results of bioassay analyses. These data were routinely recorded during the operation of a uranium hexafluoride feed manufacturing facility by the ORGDP wherein personnel are subject to relatively low concentrations of both soluble and insoluble normal uranium compounds. The study covers a two-year period during which time approximately 500 spot-air samples, 4000 shift-length air samples and 700 urine specimens were collected.

Statistical analyses of these data showed no significant correlation between either the short-term or long-term air-borne uranium concentrations, and the employees' urinary excretion rates. However, despite the many variables introduced by actual field conditions as contrasted with a controlled laboratory study, there was a statistically significant, positive correlation between the average uranium α-surface activity and the average urinary excretion rate for the individuals routinely assigned to the feed plant. A similar relationship is also evident in the comparable data obtained during an 18-month period involving personnel working in an area having low levels of soluble air-borne uranium in a sampling facility handling UF_6 enriched in the U^{235} isotope and employees subject to both soluble and insoluble uranium compounds with varying degrees of enrichment in the equipment cleaning facility.

The relationship shown may thus provide a convenient method for estimating the potential exposure for groups of employees assigned routinely to operations wherein varying concentrations of uranium compounds are found. The uranium surface contamination levels can rapidly be measured and provide an accuracy within about one order of magnitude of the predicted values for the average personnel exposure.

The post-nuclear attack water contamination problem. W. J. LACY and M. J. STANGLER, Office of Civil Defense Mobilization.

Increased attention has been focused on the post-attack problems because of an increasing concern over National security by individuals and various governmental agencies, together with an increased knowledge of the potential hazards due to fallout from nuclear weapons.

The estimation of the probable levels of contamination to be expected in public water supplies is one of the problems considered. Under fallout conditions contaminated water is not as great a hazard as the external γ-dosage one may receive simultaneously. Water which a householder may be able to get out of his water tap will in all likelihood, if from a surface source, be contaminated. This paper discusses contamination conditions, fallout levels, internal dose criteria, water contamination producing particular doses to the lower large intestine and the early radioiodine ingestion problem.

Calculations indicate that the problem of a fallout-contaminated water supply will not be severe in most cases. Radioiodine concentration by the thyroid and the resultant exposure is more serious the first 100 days after attack than the dose to the bone. However, the radiation exposures are such that they and the water supply hazard should not be ignored. Radiation detection instruments will be needed to measure the radiation from the water supply to determine the degree of contamination present.

An evaluation of the health physics problems from thorium and its daughters in a thorium purification and fabrication process. C. M. WEST, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

An adequate evaluation of the health physics problems associated with thorium processing is considerably complicated by the presence of the short-lived thorium daughter products from the decay chain. This complication was noted during extensive health physics coverage of activities which involved purification and fabrication of thorium metal. This process, including such steps as arc melting, machining, rolling, hammering, and hydro-forming, was monitored by analyses of air, smear, process materials, and direct radiation measurements. It was found necessary to give special attention to the effects of the daughters in the thorium series in the interpretation of potential health hazards. This was done by α- and γ-spectrometry and chemical analyses in addition to the traditional methods of beta and alpha counting. *In vivo* γ-spectrum measurements were made on persons involved in the process. Typical and illustrative data are reported.

Data gathered during a thorium fire indicate that more than 95 per cent of the α-activity on the original count can be attributed to isotopes other than thorium. Data from a machining operation indicate that 90 per cent of the α-activity on a sample counted two hours after collection was due to Rn^{220} daughters. It is concluded that:

(1) A realistic evaluation of the health physics situation by means of the traditional counting method of analyses is laborious and involves extended time periods before a final comprehensive conclusion can be reached.

(2) *In vivo* γ-counting appears to be the best available method for estimating the lung burden of personnel exposed to insoluble compounds of thorium.

(3) Daughter product activity may, in some cases, define the limiting potential exposure conditions.

Radiological protective apparel program at Hanford. T. C. MEHAS, Hanford Atomic Products Operation, Richland, Washington.

The seemingly diverse applications for which protective apparel are required as well as the wide range and choice of wearing schemes available may lead to poor selection or poor utilization of apparel. The result may be inadequate protection, high cost, and poor personnel considerations. The Hanford Atomic Products Operation embarked some years ago on a program to establish sound protective clothing practices by considering all logistic factors from initial procurement to ultimate utilization. The development and incorporation of this program as well as the many benefits derived from it are discussed. The principal topics of discussion include the development and acceptability of radiological apparel, preparation and utilization of standards for procurement of apparel, control and dispersal techniques, maintenance, and the overall economic impact of the program.

Criteria for appropriate establishment of removable radioactive surface contamination limits. G. W. SPANGLER, Atomics International, Canoga Park, California.

An experimental and theoretical determination of the potential radiological hazards that may be incurred from various levels of removable radioactive surface contamination is described. Major cost considerations are discussed, which combined with the potential hazard analysis provide a means whereby appropriate contamination limits may be chosen.

Internal emitters—II. Whole Body Counting Studies. Chairman: W. J. BAIR, Hanford Atomic Products Operation, Richland, Washington.

Quantitative measurements of some γ-ray emitting radionuclides in nuclear industrial workers by Whole Body Counting techniques. F. SWANBERG, JR., Hanford Atomic Products Operation, Richland, Washington.

A summary and an analysis of the potassium-40, zinc-65 and cesium-137 detected and measured by Whole Body Counting methods in more than one thousand persons during 1960 are presented. These data are the result of a wide sampling of the employees in a large nuclear industrial center. Workers representing all crafts and skills are included. Distributions of the quantitative results and estimates of the mean value for the nuclear industrial population are presented.

Problems and methods of determining zinc-65 in humans. H. E. PALMER and W. C. ROESCH, Hanford Atomic Products Operation, Richland, Washington.

Zn^{65} does not immediately attain its final distribution in the body. It is initially deposited in the central part of the body and then slowly distributes through the rest of the body during the next 200 days. Because of the changing distribution it is impossible to calibrate accurately the inclined chair method for Zn^{65}. A scanning method is described which is much less dependent on the isotope distribution than the chair method. This method is good for measuring all gamma emitting isotopes regardless of the position of the isotope in the body. The retention and distribution of Zn^{65} in the body is discussed. Further applications of the scanning counter are also described.

The Whole Body Counting of persons involved in SL-1 reactor criticality incident of 3 January, 1961. L. L. SKOLIL, J. I. ANDERSON and A. HARBERTSON, U.S.A.E.C., Health and Safety Division, National Reactor Test Station, Idaho Falls, Idaho.

Sixteen persons who were involved in the operations after the SL-1 reactor criticality incident (3 January, 1961) at the National Reactor Testing Station, are being studied in the Whole Body Counter. Iodine-131 was observed as the chief initial body burden. Cesium-137 was observed in most of these persons after iodine was eliminated. The maximum body burden for Iodine-131 in any of those observed did not exceed 1 μc. The effective decay rates of iodine and cesium are being determined from whole-body counting.

The Whole Body Counting of persons involved in silver-110 incident at the National Reactor Test Station. L. L. SKOLIL, J. I. ANDERSON and A. HARBERTSON, U.S.A.E.C., Health and Safety Division, National Reactor Test Station, Idaho Falls, Idaho.

Over fifty persons who received body burdens of silver-110 resulting from air-borne activity at the ETR reactor at the National Reactor Testing Station on 7 January, 1961, are being studied. The maximum body burden in all cases was less than 0.5 μc. During clean-up operations re-exposure did occur. An effective half-life of approximately 8 days was determined for some of those receiving sufficient body burden for decay studies. Analysis of urine and fecal samples indicate that the elimination proceeds via fecal extractions.

Bioassay Problems. Chairman: R. G. THOMAS, Lovelace Foundation, Albuquerque, New Mexico.

***In vivo* γ-spectrometry for inhalations of cobalt-60, neptunium-237–protoactinium-233, and zirconium 95–niobium-95.** R. E. COFIELD, Y-12 Plant, Union Carbide Nuclear Company, Oak Ridge, Tennessee.

Techniques, calibrations, and efficiencies are reported for the measurement of the amount of three radioisotope systems in the human lungs. The measurement technology for Co^{60}, $Np^{237} + Pa^{233}$, and $Zr^{95} + Nb^{95}$ inhalations is based on *in vivo* γ-spectrometry with a large (9 in. diameter $\times$ 4 in. thick) NaI scintillation detector in an "iron room" type low background counting chamber. The limits of detection are 0.00026 μc of Np^{237} in equilibrium with Pa^{233}, 0.0006 μc Co^{60}, and 0.0014 μc of Zr^{95}, or Nb^{95} or $Zr^{95} + Nb^{95}$. These levels of detection

are all less than 2 per cent of the amount of the radioisotopes which would deliver 0.3 rem/week to the lungs. Data from repetitive measurements on actual inhalation cases are presented for Co^{60} and $Zr^{95} + Nb^{95}$.

Accidental personnel exposure to elemental sulphur-35. A. R. MAASS, T. L. FLANAGAN, D. BLACKBURN and M. SMYTH, Smith, Kline and French Laboratories, Philadelphia, Pa.

In March 1960, an employee was contaminated externally and internally following explosion of an ampule containing 1 c of elemental S^{35} in benzene. No explanation has been found for the explosion, but precautions taken to prevent a recurrence will be discussed. Decontamination of the laboratory was accomplished without incident; the procedure will be outlined.

Steps were taken to estimate and reduce personal body burden. Multiple blood, urine and fecal samples were obtained within 1 hour after the accident. Sampling was continued for 1½ weeks, until no further radioactivity could be detected. Plasma radioactivity exhibited a biological half-life of 19 hr. Eighty-five per cent of total urinary radioactivity and 90 per cent of total fecal radioactivity were excreted within 24 hr. Body burden was estimated to be 25 μc. Observations will be discussed based on data of S^{35} metabolism.

PUQFUA: An IBM-704 FORTRAN code for determining plutonium body burden from urine assays. J. N. P. LAWRENCE, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

PUQFUA is an IBM-704 FORTRAN program, written primarily for Pu^{239}, which calculates from urinalyses the plutonium body burden in microcuries and the per cent of the maximum permissible body burden. It is based on a set of power function elimination equations developed by WRIGHT H. LANGHAM for the excretion of plutonium over a five-year period. Results with PUQFUA compare favorably with those of other methods. In general, it is easy to apply for a large number of individual samples, and the validation method is objective and requires no past experience with plutonium body burden calculations on the part of the evaluator.

Studies on rates of elimination of tritium-labeled water in humans. R. MOORE and E. R. BUSKIRK, American National Red Cross, Washington, D.C.

Tritium-labeled water was injected intravenously and the time course of specific radioactivity in plasma, red cells, urine, and expired water vapor was determined and compared. Specific activity of breath water and of urine followed a pattern of damped oscillations about the curve for the specific activity of plasma water. The transfer function of the biological system was determined for the injection of radioactivity. Specific radioactivity in body fluids was described by a sum of three or more exponential terms. These results were used to recalculate the whole-body radiation exposure due to tritium-labeled water. Results indicated that breath water samples may, under certain circumstances, be substituted for urine or plasma water samples in the determination of body burden of tritiated water.

Infant retention studies. C. P. STRAUB and N. C. TELLES, Rbt. A. Taft Sanitary Engineering Center, Public Health Service, Cincinnati, Ohio.

Studies were initiated in July 1960 to determine the retention of strontium-90, other radionuclides, and stable elements in six infants ranging from 7 to 26 weeks in age. They were fed commercially available baby foods *ad libitum*. Food, water, and formula intake were carefully measured. All excreta (feces and urine) were collected in disposable diapers and the strontium-90 content measured. Data have been composited over periods of 4 weeks and the difference between intake and excreta levels used to provide the basis for calculating retention of the specific substance of interest.

Procedural details are spelled out and preliminary findings are reported as a function of the age of the infant.

Surface Contamination and Environmental Radiation Measurements. Chairman: E. ALPEN, U.S. Naval Radiological Defense Laboratory, San Francisco, California. Co-Chairman: H. J. DUNSTER, United Kingdom Atomic Energy Authority, Harwell, England.

Studies of surface contamination—I. Intercomparison of methods for measuring "removable" contamination. G. W. ROYSTER, JR., and B. R. FISH, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

In a study of the measurement of surface contamination the smear and adhesive paper techniques were compared with a new method of sample collection, "smair" samples, in which loose contamination is redispersed by air impingement and the resulting air-borne material is collected on a filter paper. Nearly uniform contamination was produced on test surfaces by dispersing ThO_2 particles, 3 μ in diameter, into an enclosure and allowing them to deposit by gravity settling. Smear, adhesive paper, and "smair" samples were taken at points for which the total deposited activity was known. Adhesive paper samples showed the least variability in per cent removed from the various surfaces (54–86 per cent). Smear samples showed a wider range of removability (17–75 per cent), depending upon the porosity and hardness of the test surface. The per cent removed by the "smair" sampler, as a function of surface, varied more than did the other two types of samples (2.4–17 per cent). These observations emphasize the danger of assuming one method of removing surface contamination to be representative of any other mechanism of redispersion.

A portable dose rate instrument for measurement of natural background radiation levels. F. L. RISING, Hanford Atomic Products Operation, Richland, Washington.

An instrument of the ionization-chamber type which is

capable of measuring radiation dose rates down to and below those encountered in natural background has been designed and constructed. It consists of a 40 l. ionization chamber coupled to a Keithley Model 600 portable battery-powered electrometer. The chamber polarizing-battery is a part of the chamber center electrode assembly and is located inside the chamber.

The radiological safety program at Indian Point Station. W. F. Nelson, Consolidated Edison Company, New York, N.Y.

Indian Point Station consists of a nuclear reactor and a conventional oil fired electric generating station which will supply electricity to the New York City area. The radiological safety program is operated by the personnel in the Test Group who are also responsible for the maintenance of all instrumentation at the plant. The radiation and contamination monitoring equipment has been organized into a Health Physics Monitoring System based on the concept of individual monitoring within a controlled area and environmental monitoring beyond this area. Additional radiation monitoring is furnished by the plant functional monitoring equipment which detects the presence of radioactive contamination in any of the auxiliary systems.

Surface contamination measurements as an index of control of radioactive materials. H. J. Dunster, United Kingdom Atomic Energy Authority, Harwell, England.

The purpose and the objections to surface contamination measurements will be reviewed. The need for permissible values in this field is evident and the general basis of the derivation of the values in use in the United Kingdom will be discussed. Surface contamination can provide an indicator of the degree of control which is being maintained over operations with radioactive materials and, in most cases, is a reliable indication of whether air monitoring is required.

Limitations of commercially available instrumentation for analysis of environmental media. R. K. Stoms, R. Brueneman and J. Berger, Public Health Service, Cincinnati, Ohio.

Methods and equipment concerning measurements of low level γ-emitting nuclides are discussed. Low count rates result in problems in obtaining accurate, reliable data.

Satisfactory results are based on selection of equipment, calibration, sample preparation, amount of radioactive contamination in reagents, and the minimization of background and interfering nuclides.

The problems of using instrumentation which is basically designed for higher level counting are defined. Selection and use of this equipment are discussed and various calibration methods compared. Sample size and preparation and possible radioactive contamination of reagents are related to counting results. Methods are given to minimize background and the interference of radionuclides.

X-radiation surveys of microwave generators. W. P. Knox, M. M. Weiss and J. D. Struthers, Bell Telephone Laboratories, Whippany, New Jersey.

The demand for more powerful microwave radar and communication systems has created a serious ionizing radiation problem for operating and maintenance personnel. The hazard arises from the generation of X-rays by vacuum tubes operating at potentials above approximately 15 kV. To determine the magnitude of the X-radiation in the vicinity of the high powered components of the system requires special instrumentation and techniques. This paper will discuss the procedures used in making radiation surveys at typical microwave generator sites and will present the results of the surveys and the safety recommendations made for these locations. The relative merits of the techniques and instruments employed in these surveys will be examined.

Improvements in air sampling techniques and equipment using membrane filters. K. J. Schlager, T. M. Allen, S. B. Gerber, G. E. Kinsella, F. J. Krupka and D. W. Reilly, Argonne National Laboratory, Lemont, Illinois.

Air sampling as a tool for evaluation of process and ventilation control, filter efficiency, etc., is becoming an important adjunct to health protection operations. Membrane filter samples provide a versatile and convenient method of collecting and analyzing such samples, for which techniques and equipment have been recently improved at Argonne National Laboratory.

Routine use of membrane filters has been simplified by the design of a very small and inexpensive filter holder, providing some advantages over other available units, e.g. reduced size and weight, tight sealing against any filter material without the twist-lock action which often damages membrane filters, and parts exposed to contamination are expendable.

Simplified techniques for preparing and counting membrane filters have been devised for use with gas-flow, internal proportional counters. A combination α-γ-scintillation counter has also been designed and constructed for routine radiation monitoring applications.

Current applications of the method, both in general laboratories and in special facilities, are described.

The radiation protection program for a large private utility power reactor. J. H. Hughes, Dresden Nuclear Power Station, Joliet, Illinois.

The over-all program for the protection of personnel on the Dresden Nuclear Power Station property and the inhabitants of its environs from harmful quantities of radiation and/or radioactive material contamination is a composite of programs developed at such installations as Hanford, Knolls and Valecitos Laboratories which are operated by the General Electric Company. Assistance in the formulation of this program by General Electric was extended as a part of contractual arrangements for the construction of the plant. The program was initiated in October 1959 by Commonwealth Edison Company's Radiation Protection Engineer.

The program is subdivided into personnel, plant, and environs monitoring portions. Areas of these subdivisions are carried on by contracts for specific services. The interdependence of the three portions of the program is borne out by the fact that results obtained thus far confirm both the adequacy of the program and that the plant performs with the original design criteria.

Discussion and description of radiation exposure experience, plant radiation levels at various power levels and shut down, and significant results of the environs survey data for the pre-operational period will be presented.

Radiation measurements over simulated plane sources. F. J. DAVIS and P. W. REINHARDT, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

To simulate an infinite plane source an array was laid out with positions at 100-ft intervals in the form of a square 2000 ft on a side. To facilitate ground measurements a smaller square array of 100 positions at 10-ft intervals was placed in the center of the large array. Two sets of sources were used in the array. Co^{60} and Cs^{137} sources of 4.5 mc and 15 mc, respectively, were available to be placed in the large array with sources one-hundreth these values in the small array. Measurements 3 ft above ground indicated 114–136 μr/hr for Co^{60} and 122–147 μr/hr for Cs^{137}; the spread of readings was due to the variation among instruments used. Measurements were made in aircraft at 100-ft intervals above the array using scintillation counters and a 256-channel scintillation spectrometer. Flights were also made over point sources of Co^{60}, Cs^{137}, and I^{131}.

Results of the "ORPHEE" and "EURIDYCE" experiment. F. DUHAMEL, J. PRADEL and M. GRAS, Commissariat l'Energie Atomique, Saclay, Paris, France.

"ORPHEE" is a special device designed for the in-pile studies of the pollution produced by non-canned uranium. "EURIDYCE" is the similar device located outside the pile. With the help of these two devices, we study the production, propagation and deposition of fission products as also the efficiency of filters and the decontamination of some circuits.

Tests began in 1959. The communication gives the first results obtained.

A summary of radiation exposures received by workers in medical isotope laboratories and X-ray departments from 1950–1960. C. K. SPALDING and R. F. COWING, New England Deaconess Hospital, Boston, Massachusetts.

Exposure to ionizing radiation received by workers in isotope and X-ray laboratories over the past ten years has not appreciably changed even though the amounts of radioactive material used and the number of X-ray films taken have increased considerably.

Data collected after monitoring a total of 1200 persons for varying periods of time show that 4 per cent of the X-ray personnel experience exposure above 100 mr/week, but only 0.5 per cent of the isotope workers receive an excess of the tolerance dose.

The reasons for these exposures have been tabulated and present a definite need for education and understanding of health physics problems by technical personnel in both fields.

Waste Disposal. Chairman: C. P. STRAUB, Radiological Health Research Activities, Rbt. A. Taft Sanitary Engineering Center, Cincinnati, Ohio.

The importance of verifying design, calculations, and assumptions with measurements, in a stack monitoring field-monitoring hazards control program. L. D. VAN VLECK, General Electric Company, Idaho Falls, Idaho.

Any stack monitoring field monitoring program depends for accuracy on the nearness of design calculations and assumptions to their true values. Several serious departures from design conditions in stack flow rates, coupled with erroneous assumptions for the collection efficiencies of high efficiency filter papers, and for the line loss parameters combine to introduce errors amounting to a factor of 5–7 in the determination of the gross fission produce stack release rate. The error for certain specific isotopes may be as high as a factor of 16–20. The assumption that the Sutton diffusion equation is adequate to describe the dispersion of radioactive material in the atmosphere appears generally sound and conservative.

Calculation of the effective height of a plume. M. LEQUINIO, Commissariat l'Energie Atomique, Saclay, Paris, France.

Various theories allow to estimate the actual height of a radioactive plume of a stack. Application of these various theories to some particular cases gives very diversified heights. Studies have been made to look for a better way of calculation. These are studies which are explained in this communication.

Health physics consideration for the disposal of non-enriched uranium chips by burning. M. SANDERS, Y-12 Plant, Union Carbide Nuclear Company, Oak Ridge, Tennessee.

A safe and economical means of reducing a radioactive, pyrophoric material to a non-pyrophoric state can be accomplished by burning of non-enriched uranium metal chips in an open pit. The pit, used for combustion of the material, provides excellent long-term storage without a significant health problem. A series of tests were conducted in an isolated area with quantities of uranium during different atmospheric conditions to determine the contribution of the operation to the levels of air-borne contamination.

Strategically located high volume air monitors reveal that the disposal of non-enriched uranium metal chips by controlled burning in an isolated area would be an acceptable practice and well within the limit as stipulated by the National Committee on Radiation Protection.

Routine measurements of ground water velocity using sulfur-35. W. F. MERRITT, Atomic Energy of Canada, Ltd., Chalk River, Ontario.

A method is described for the measurement of the effective velocity of ground water movement using sulfur-35 as a tracer. The method uses modified porous bronze piezometers for both injection of the tracer and sampling of the ground water. Sulfur-35 has been used routinely at Chalk River with good results, and a comparison of it with tritium and fluorescein dye is given. Among the advantages of sulfur-35 as a tracer is the fact that it can be detected by simple chemical and counting methods at concentrations well below maximum permissible drinking water tolerances. The usefulness of the techniques for other problems involving ground water is also discussed.

Columbia River continuous monitoring system. T. C. MEHAS, Hanford Atomic Products Operation, Richland, Washington.

The environmental monitoring program at Hanford recently included an automatic facility which continuously monitors river surface γ-activity and collects samples of Columbia River water for isotopic analyses. This paper describes the monitoring portion of that facility. It discusses the equipment and methods utilized to instantaneously detect and record γ-dose rates in the μr/hr region, total and incremental dose and differentiation between two broad energy bands. An alarm system which immediately signals any abnormal activity or any instrument's malfunction is also briefly described as is the control circuitry of an emergency sampling system which automatically provides grab-type samples of water coincidental with abnormal activity events.

Sodium attapulgite clay as a radioactive waste disposal agent. F. SICILIO, N. H. HORTON and R. J. KLETT, Georgia Institute of Technology and the Floridin Company, Tallahassee, Florida.

Conversion of liquid radioactive wastes to solids should tend to decrease the over-all cost of waste storage and disposal, with an increase in safety. Waste solutions of low salt content can be converted into an activity-free liquid stream and a solid concentrate of high activity through sorption by a solid.

The exchange-type capacity of attapulgite clay converted to the sodium form has been determined. Natural attapulgite, or Florida fullers earth, is a hydrous aluminum magnesium silicate belonging to the polygorskite group of clay minerals. It is mined by the Floridin Company of Tallahassee, Florida and it is available in granular form. Batch and percolation-type contact with five cations and one cation mixture indicate an average capacity in the order of 70 mequiv/100 g of converted clay, or about a three-fold increase over the natural product.

Leaching of loaded attapulgite with solutions of nominal acidity or basicity, after firing at 2000 F, results in a loss of only traces of the sorbed activity.

Mineral reactions—a new waste decontamination process. J. F. HONSTEAD, L. L. AMES and J. L. NELSON, Hanford Atomic Products Operation, Richland, Washington.

The assurance of safe containment to permit decay is the objective of radioactive waste fixation processes. The evaluation of the ease of managing, controlling, and monitoring such processes is of concern to the health physicist. As a fixation matrix certain minerals promise long-term stability, even when exposed to radiation and high temperatures. A review of progress in mineral reaction research at Hanford is presented.

Study of the retention of radioisotopes on mineral constituents of materials underlying Hanford ground disposal sites was broadened to include other mineral reactions suitable for decontamination of aqueous wastes. One of these is the selective absorption of a radioactive waste constituent on a mineral surface having a particular affinity for it. The preferential absorption of cesium from solutions of sodium by the natural zeolite clinoptilolite is an example. Another type of reaction involves the gradual replacement of a mineral by another form as a solution containing the replacement ion passes through the bed. Radioactive ions that fit into the new mineral lattice are concurrently removed from solution by being thus fixed in the solid. For example, the replacement of calcite (limestone) by apatite (similar to bone mineral) as phosphate solution passes through the bed results in the simultaneous removal of bone seeking radioisotopes from solution. Effects of operating variables and the decontamination capabilities of some of these reactions are presented.

The feasibility of applying mineral fixation to Hanford high-level waste was investigated. Results are presented from some laboratory studies with samples of neutralized waste and beds of clinoptilolite. Included are preliminary indications of bed capacities and possible ways of incorporating mineral reactions into other fixation schemes.

It is concluded that mineral reactions show considerable promise as means of immobilizing radioisotopes from aqueous wastes. They do not represent a panacea for all waste disposal problems but are an addition to the array of available processes from which waste decontamination methods are selected.

Cesium exchange properties of vermiculite. D. G. JACOBS, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Although seepage pits effectively decontaminate the intermediate-level waste stream at ORNL, there is concern regarding the lack of control of the effluent quality once the wastes have been discharged to the pits. The use of a mineral-filled column is regarded as an inexpensive, yet efficient, alternative method for waste decontamination.

For this purpose the cesium exchange properties of various grades of commercially available vermiculite were investigated and compared to other natural ion-exchange materials. The distribution factors (K_d) obtained for the

vermiculites in a system similar to the ORNL intermediate-level waste stream were not as high as those obtained for some of the collapsed (10 Å), 2:1 layer lattice minerals, but the sorptive properties of vermiculite were found to be quite adequate for practical application; furthermore, the physical properties were far superior to other clay materials.

A cesium K_d of 70 ml/g was obtained for natural vermiculite after slurrying 24 hr, increasing to 193 ml/g after 500-hr contact time and to 220 ml/g after 1000 hr. The sorptive properties were markedly improved by subjecting the vermiculite to potassium treatment or to heat treatment.

A tenfold decrease in the stable ion composition of the waste solution resulted in a tenfold increase in K_d, as predicted from mass action. This permits extrapolation of the data for consideration in the use of vermiculite columns for decontaminating other waste streams, such as the ORNL process wastes.

Data obtained by bench-scale and field-scale (10 ft long, 2 in. inside diameter) column studies compare closely with those obtained by slurry studies. The breakthrough curves obtained were sufficiently steep to indicate that the partial cesium sorptive capacity of a vermiculite column could be completely utilized if two columns were operated in series. A column 10 ft long and 10 ft in diameter filled with Na-treated vermiculite is estimated to be capable of decontaminating more than 5×10^5 gal of ORNL intermediate level waste.

Processing by resins of radioactive liquid effluents of intermediate level. M. COHEN, Commissariat l'Energie Atomique, Saclay, Paris, France.

The possibility to purify waters by ion exchange resins is well known, but had not been until now the object of an industrial processing on the effluents from a nuclear plant. These are researches made in view of this industrial processing which are explained in this communication.

Significant results in low-level waste treatment at Oak Ridge National Laboratory. K. E. COWSER, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Operating experience with the 500,000 gal/day waste treatment plant at ORNL has demonstrated that it exceeds the design specifications, i.e. removal efficiency of greater than 50 per cent. An average of 85–90 per cent of the Sr^{90} and Cs^{137} are removed. There is nevertheless, a distinct limitation on the maximum efficiency obtainable with such a single-stage, chemical precipitation treatment system. The full potential of the treatment plant can be realized only by modifying the process or by adding to the facility.

A number of experiments were performed, under controlled laboratory conditions, to investigate the following: the combination of lime, soda ash, and clay that would remove the greatest amounts of Sr^{90} and Cs^{137}; the use of phosphate as a primary coagulant and as a supplement to lime-soda softening for Sr^{90} removal; the addition of coagulant aids to increase the removal of suspended solids that are associated with 50 per cent of the effluent activity; and the application of vermiculite and rock phosphate columns for second-stage treatment.

The optimum combination of primary coagulants was established, while the addition of phosphate as a supplement to normal chemical feed increased the removal of Sr^{90} to 98 per cent. Coagulant aids improved the settling characteristics of the flocculent material, while vermiculite exhibited distribution coefficients for Sr^{90} and Cs^{137} in excess of 5000. Process changes or possible additions to the plant have been evaluated by pilot scale tests or by direct application to the operating system.

Strontium removal from waste solutions. T. TAMURA, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

For possible use as sorbents in waste treatment columns, the abilities of different minerals and compounds to selectively remove strontium from 0.1 M $NaNO_3$ solutions were compared. Slurry tests showed that strontium removals which depended on ion exchange reaction were only slightly affected by varying the pH between 5 and 10. Materials such as alumina which depended on their surface characteristics were most sensitive to pH changes and were adversely affected by low pH. When orthophosphates (100–5000 p.p.m. PO_4) were added to phosphatic minerals or to compounds capable of forming insoluble phosphates, strontium removals were generally increased. Addition of orthophosphates to systems involving ion exchange reactions for the removal of strontium resulted in an increase or decrease depending on the exchangeable ion on the material. Thus, for calcium or magnesium saturated vermiculite addition of Na_2HPO_4 increased removal; for sodium vermiculite the addition of 100 p.p.m. phosphate decreased removal. The influence of the structural aspects was found to be significant; potassium saturation on vermiculite collapsed the layer-lattice structure and reduced the strontium exchange reaction. The selectivity of commercial grade materials such as rock phosphate, coconut charcoal, bone charcoal, vermiculite and resins was investigated.

Dosimetry. Chairman: W. S. SNYDER, Oak Ridge National Laboratory, Oak Ridge, Tennessee. Co-Chairman: L. S. TAYLOR, National Bureau of Standards, Washington, D.C.

Radiation yield from some bare critical assemblies. R. H. RITCHIE, H. B. ELDRIDGE and V. E. ANDERSON, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Methods of calculating the radiation yield of critical nuclear assemblies are of great importance in assessing the amount of ionizing radiation received by persons exposed in criticality accidents. Calculations of the leakage from some bare assemblies have been carried out, using a straightforward approach, and have been compared with experimental data obtained by members of the Health Physics Division, ORNL. It is found that the approach gives reasonable accuracy for distances from the reactor which are usually of interest in criticality accident studies.

Application of phantoms to studies of blood sodium activation by neutrons. J. A. AUXIER, F. W. SANDERS and W. S. SNYDER, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

The production of Na^{24} in the human body by neutron irradiation is an important part of a system of monitoring nuclear accidents. Feasibility studies have been made of phantoms filled with homogeneous aqueous solutions of sodium for the application of sodium activation to dosimetric investigations of nuclear accidents. The Na^{24} production per unit neutron dose was measured in anthropomorphous phantoms, cylinders, and live burros.

The quantity of Na^{23} activated to Na^{24} was found to be a function of body size and orientation relative to the source, but the analysis was found to be negligibly influenced by blood sampling time in the interval 2–24 hr post-irradiation. Thus, the biological half life is relatively long compared to the radioactive half life and the sodium exchange between blood and other tissues is, at least approximately, in equilibrium during the 2–24 hr interval.

Sodium activation in phantoms has also been computed by the Monte Carlo technique for several neutron energies.

Casuality assessment device for measuring neutron-induced body activity. E. R. BALLINGER, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

A portable NaI crystal photomultiplier assembly calibrated to read the incident radiation of neutrons required to produce the body activity observed when corrected for neutron energy estimates by means of copper foils is described.

Some convenient representations for fission neutron distributions in air. R. L. FRENCH, Convair, Fort Worth, Texas.

An analysis of Monte Carlo results for fission neutron scattering in air has revealed that, for slant ranges of 500–2000 yards, the angular distribution (dose per unit solid angle as a function of angle β measured from the source-receiver axis) of the scattered neutron dose from a point isotropic source may be approximated by

$$\frac{dD_2}{d\Omega} = a + be^{-c\beta}$$

This expression, coupled with an exponential expression for the total scattered dose as a function of range, permits a simple and convenient approach to many dosimetry and shielding problems which require detailed angular and spatial distribution data. The validity of the Monte Carlo data considered in the analysis has been demonstrated by comparison with measured data from weapons tests.

A comparison of some recent calculations of neutron transport in air. M. B. WELLS, Convair, Fort Worth, Texas.

Monte Carlo computations performed by J. I. MARCUM of the Rand Corporation, W. A. BIGGERS of the Los Alamos Scientific Laboratory, C. R. MEHL of the Sandia Corporation, and M. B. WELLS of Convair, Fort Worth, to determine the transport of 3- and 14-MeV neutrons in air and air–ground are compared and the differences between these computations are discussed.

Air-transport calculations were performed by WELLS, who used the neutron cross sections given by BIGGERS as cross section input data. WELLS' results are found to be in good agreement with BIGGERS' air–ground calculations, but disagree with MEHL's calculations for air only.

Monte Carlo calculations of the transport of neutrons from a point isotropic fission source in air, as made by WELLS, are compared with results obtained from a moments-method calculation, performed by D. SPIELBERG at the Nuclear Development Corporation of America. The differences between these calculations are discussed.

An X-ray source for energy dependence studies in the 10–100 keV range. R. SANNA and A. SHAMBON, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York City.

An X-ray facility has been established for the purpose of determining the degree of energy dependence of radiation instrumentation which has been previously calibrated with cobalt-60 and radium sources. Of special interest is the response of environmental radiation survey instrumentation in the low energy region. Secondary X-rays from the *K* shells of copper, cadmium, lead and uranium are utilized for this work. A standard air chamber for the determination of the absolute value of the dose rate at points of interest. All the equipment, with the exception of a standard air chamber, is commercially available.

Preliminary measurements indicate that the secondary beam is of the expected energy and purity. Calibrations of the *K* sources and energy dependence studies of existing instrumentation have been obtained.

Scattered radiation from large cobalt-60 calibrating sources. L. COSTRELL and H. O. WYCKOFF, National Bureau of Standards, Washington, D.C. (L. S. Taylor presenting paper).

Radiation beams from multicurie cobalt-60 sources are widely used for instrument calibration and for radiation treatment. It is often assumed that such beams contain only the 1.17 and 1.33 MeV γ-rays. However, it is recognized that this assumption is not strictly valid because of the energy degradation due to Compton scattering in the source material and the source collimator. A scintillation spectrometer with a 5 in. diameter by 4 in. long sodium iodide crystal has been used to determine the amount and energy of the scattered radiation as well as its variation with source dimensions and collimator geometry. Since the sources currently in common use have appreciable mass, source scattering is a far greater contributor of degraded radiation than is the collimator. Quantitative data are presented.

A developer modification to increase the range of dosimeter films. H. M. CLEARE and N. H. KING, Eastman Kodak Company, Rochester, New York.

The useful range of photographic dosimeter systems may be increased significantly by a simple modification of

the developer. Dosimeter films, processed in a developer to which sugar has been added, show a preferential loss in sensitivity at high densities. The sugar lowers the diffusion rate of the developing solution into the emulsion, thereby producing a local exhaustion effect in high density areas.

As an example, the sensitive component of Kodak Special Personal Monitoring Film, Type 3, processed for 5 min in Kodak Rapid X-ray Developer containing 200 g/l. of sugar, shows no loss in sensitivity below a net density of about 0.3 when compared to 3 min development in the same developer but without the sugar. At higher densities, the film becomes progressively less sensitive than when processed in the normal developer. At a density of 5.0, the film processed in the "sugared" developer is about 15 times less sensitive than when processed normally.

A miniature 2π dosimeter. D. E. HEGBERG, Hanford Atomic Products Operation, Richland, Washington.

Using the thermoluminescent phosphor CaF_2–Mn as a detector, a small β–γ-dosimeter has been developed. The sensitivity of the dosimeter is such that doses from 1.0 mr to 10 r can be measured with an accuracy of 10 per cent if read-out is effected immediately after an exposure period of not greater than 8 hr. The device is energy independent, to ± 10 per cent, from 18 keV to several MeV and dose rate independent for dose rates less than 20 r/hr. The device is essentially adapted to a 2π geometry. More specifically, the dosimeter has been fabricated as an experimental finger-ring dosimeter to be used principally by plutonium metal workers. The dosimeter materials are inexpensive, and the cost of fabrication is reasonable. The read-out instrumentation consists of an induction heater, an amplifier–chart recorder, and photomultipliers.

Current developments in the use of nitrous oxide in the radiation dosimetry associated with the Lockheed Radiation Effects Reactor. R. E. SIMPSON, Lockheed Aircraft Corporation, Marietta, Georgia.

When exposed to a source of ionizing radiation of doses ranging from 10^6 to 10^{10} ergs per gram-carbon nitrous oxide decomposes linearly as a function of the absorbed dose in the ratio N_2:O_2:NO_2:1.0:0.14:0.48 as stable products. With the addition of a small amount of U^{235} to the dosimeter, the gas can be used in neutron dosimetry.

The principle of HARTECH and DONDES (*Nucleonics*, p. 66, March 1952) has been utilized in the development of nitrous oxide in the dosimetry program associated with the Lockheed Radiation Effects Reactor (LRER). This has included the design of practical dosimeters and vacuum analytical apparatus, equipment calibration and the development of a routine method.

Prior to use at the reactor the dosimeters were calibrated in the pure γ-field of the 23,000-c cobalt-60 source of WADC using a 4 cm^3 graphite ionization chamber as a basic reference standard. Gaseous decomposition values were expressed in terms of mole-fractions of nitrogen–oxygen to nitrous oxide. With a satisfactory γ-calibration the dosimeters were then exposed to the mixed radiation field of the LRER to obtain a reactor calibration. This included the selection of locations of definitely known γ-doses under varying reactor power levels and shielding configurations. This produced a definite linear relation between reactor power and γ-dose at any other location.

Although the reactor exposure data produced a similar curve as the pure γ-data, there are definite indications of the neutron effect in the decomposition values of the gas that bias the γ-dose. However, the expected break in the decomposition curve is obtained at the γ-equivalent dose of 10^{11} ergs per gram-carbon. The neutron effects are being further investigated (F. MOSELEY and A. E. TRUSWELL, AERE-R3078, United Kingdom (1960).

The advantages of nitrous oxide as a total dose dosimeter include post-irradiation stability, flexibility and economy. It is adaptable to a variety of uses including high flux material testing and total dose monitoring in areas of high radiation exposure.

Radiobiology and Ecology. Chairman: S. P. HUNGATE, Hanford Atomic Products Operation, Richland, Washington. Co-Chairman: H. V. LARSON, Hanford Atomic Products Operation, Richland, Washington.

Modification of zinc-65 absorption by dietary zinc. C. R. RICHMOND and J. E. FURCHNER, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

An attempt to modify the absorption of oral Zn^{65} in rats was made by the addition of natural zinc acetate to the diet of male Sprague-Dawley rats. After the rats were maintained on a zinc-supplemented diet for 4 weeks, an oral dose of $Zn^{65}Cl_2$ was administered to the rats. The retention of Zn^{65} was determined by whole-body assay techniques. Differences in per cent Zn^{65} retained were attributed to differences in the zinc acetate level in the diet.

Data from 1 human subject suggest that intestinal absorption may be decreased by increasing dietary zinc levels.

Bioaccumulation of radioactive isotopes by herbivorous small mammals. S. V. KAYE and T. B. DUNAWAY, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Sr^{90}, Ru^{106}, Cs^{137}, and Co^{60}, in that order, are the most abundant radionuclides in tissues of mammals resident on the radioactive dry bed of the former White Oak Lake of the USAEC Reservation at Oak Ridge, Tennessee. Although the Sr^{90} per unit weight of soil and per unit weight of food is less than that for the other radionuclides, the Sr^{90} per unit weight of whole animal carcass is highest. These radioassays confirm theoretical calculations which show that the body burden of Sr^{90} at time t is greater than that for Ru^{106}, Cs^{137}, and Co^{60}.

Reduction of radioactivity in milk. R. W. Carter, W. V. Willis and R. H. Fetner, Engineering Experiment Station, Georgia Institute of Technology, Atlanta, Georgia.

A method to reduce radioactivity from fallout in milk has been investigated. Ten dairy animals were confined in dry-lot (removal from green pasturage) for 7 weeks and the cesium-137 content of milk samples from this group was compared to samples from a ten animal control group. Cesium-137 content was analyzed weekly on collective samples which were taken daily from each animal and pooled with the appropriate group (experimental or control). The reduction in cesium-137 content of the weekly samples varied from 27 ± 13 per cent to 60 ± 15 per cent—the average reduction over the 7-week period was 38 ± 14 per cent.

Radiological surveillance of total diets in selected locations in the United States. H. F. Kraybill, C. Clavert, W. M. Decker and J. G. Terrill, Public Health Service, Washington, D.C.

Coöperative studies by the above Divisions, through assistance of State and local health departments, have facilitated planning and initiation of broad studies on radioactivity in total diet and single foods of a selected population in the following eight regions: New York, Chicago, St. Louis, Atlanta, Dallas, Denver, Seattle and Los Angeles. Sampling of foods is at point of consumption. Criteria for selection of population and assessment of food consumption and in turn food radioactivity are age, socio-economic status, geographical location and religious or ethnic groups. Initial studies are concentrated on children—a critical group for radiation exposure, with boarding schools used for sampling. Expansion of total diet studies to radioactivity indicator foods are planned on the basis of volume of food consumed, calcium content of food and known high levels of radioactivity in certain foods. Data on food intake and concomitant dietary intake of radioactivity for groups studied are presented.

Strontium-90 monitoring at the Savannah River Plant. S. E. Butler, Savannah River Plant, Aiken, South Carolina.

Strontium-90 was determined in milk and other samples collected in the environs of the Savannah River Plant. Milk samples were prepared for analysis by absorption of Sr^{90} on cation exchange resin, a procedure which avoids lengthy evaporation and ashing of whole milk. Various procedures were employed to prepare other samples for analyses. An extraction technique was used in the final analyses for Sr^{90} in all types of samples. The concentration of the nuclide was comparable to that found in other fallout studies.

Determination of strontium-90 in sea water. H. Francois, Commissariat l'Energie Atomique, Saclay, Paris, France.

A new method of determination of the strontium–yttrium couple has been developed. It is much shorter than the old one; it is also very sensitive and allows to carry out very easily the determination in sea water.

Semi-quantitative relationship of radiation and neoplasms in man. V. E. Archer and L. Simpson, National Cancer Institute, Washington, D.C.

The hypothesis that a linear relationship exists between dose and induction of neoplasms is supported by data for humans. Under this hypothesis, gram rad dosages to groups of individuals who have been followed, is used to calculate various neoplastic doses. Infants are shown to be more prone to radiation induced neoplasms than adults. It may be concluded that even though there are uncertainties in the data and in the hypothesis, the conservative approach to the irradiation of man will utilize these figures until the basic assumptions are disproved, or until more precise figures become available.

An epidemiological study of the effects of microwaves on the lens of the eye. M. Zaret, S. Cleary and M. Eisenbud, Department of Ophthalmology and Department of Industrial Medicine, New York University, Medical Center, New York City.

Although it is known that cataractous changes can be produced in the crystalline lens of experimental animals, the literature contains no reports of studies of the eyes of personnel exposed to this kind of radiation, nor reports of the environmental radiation levels to which they are exposed.

Under the sponsorship of the U.S. Air Force, a study has been made of the changes that occur in the lenses of exposed and unexposed personnel. A standard procedure for documenting lens abnormalities has been developed, as well as a semi-quantitative procedure for characterizing the severity of post-occupational exposure.

The report describes the methods used in this study and summarizes the observations that have been made to date.

Dependence of rate of recovery from acute γ-ray exposure on size of the conditioning dose. J. S. Spaulding, T. T. Trujillo and W. L. Lestourgeon, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

A homogeneous population of 4658 RF strain female mice was used to study rate of recovery from acute Co^{60} γ-ray exposure and its dependence on the magnitude of the first dose.

The time required for RF female mice to repair one-half of the radiation damage from acute Co^{60} γ-ray exposure was 7 days, and the repair half-time was not influenced by the magnitude of the conditioning dose between a dose range of 205–692 rads.

Multivariate analysis and the IBM-704 computer applied to ABCC data on growth of surviving Hiroshima children. J. C. Nehemias, U.S. Atomic Energy Commission, Washington, D.C.

Anthropometric data on children exposed to the atomic bomb were collected by the Atomic Bomb Casualty

Commission as part of a study of long-term effects of the bomb upon human beings. These data have been analyzed, using multivariate techniques with anthropometric measurements as variables, for difference in size, growth rate, and change of growth rate among groups graded roughly according to radiation exposure received.

Significant differences were observed among the groups, at all age levels studied, according to size; and among the groups in the post-pubertal age levels, according to growth rate and change in growth rate.

Miscellaneous Topics in Health Physics. Chairman: J. R. HORAN, Idaho Operations Office, U.S.A.E.C., Idaho Falls, Idaho. Co-Chairman: W. D. BURNETT, Sandia Corporation, Albuquerque, New Mexico.

Direct underwater maintenance of the GCRE reactor. W. S. GAULT, J. M. JANIS and M. K. WU, Aerojet-General Nucleonics Co., Idaho Falls, Idaho.

The GCRE reactor is located in a pool of water 30-ft deep. After several months of operation, a severe leak of the reactor coolant gas from the pressure vessel main flange was experienced. To perform the remedial work from the work bridge over the pool would entail an extensive remote tool development program due to obstruction of the flanges by the field-routed piping, cooling manifold and instrumentation leads. It was determined feasible to have underwater workers perform the mechanical functions, using simpler remote tools at a reasonable proximity to the reactor. The radiation field surrounding the reactor was measured using emulsion films and ion-chamber type survey meters. The measured γ-radiation levels ranged from 170 r/hr to 600 r/hr at 1-ft distances from the reactor flange, and from 20 mr/hr to 70 mr/hr at 5-ft distances from the reactor flange. The remedial work was successfully accomplished by two professional "hard hat" divers in a 104 man-hour period. The highest total body exposure received by either diver during his 52-hr work period was 565 mrems. The underwater and radiological hazards, the control methods, and procedures are described in the paper.

A monitor for the prompt detection of air-borne plutonium. J. CHESTER, Brookhaven National Laboratory, Upton, New York.

A Savannah River type annular impactor has been adapted to utilize a moving zinc sulphide coated tape as the collecting surface. This has resulted in a substantial increase in sensitivity since the efficiency of α-counting can be increased greatly by impacting directly on the scintillating surface. In addition, counting is carried on continuously during the collection time thereby reducing the delay in obtaining an indication of peaks in air-borne α-activity. Mechanical design of the equipment, the sensitivity achieved and the method of calibration will be discussed.

A system for β-scintillation counting. G. A. WELFORD and R. N. DONELSON, U.S. Nuclear Corporation, Burbank, California.

An inexpensive low background system capable of efficiently determining β-activities in a variety of samples has been developed.

Backgrounds of 2–3 c.p.m. are maintained and the β-energy from Y^{90} mounted as the oxalate, with plastic backing is counted at 40 per cent efficiency. The sample or source is mounted on the conventional ring and disc and covered with thin Mylar film. The detector, Pilot Scintillator B, prepared in 1-in. discs of 10 mils thickness, is secured to the Mylar with a light coating of silicone fluid. This assembly is placed directly on the face of a photomultiplier tube which is spring loaded in a U.S. Nuclear SPIA Scintillation Probe. The output from the photomultiplier is amplified within the probe and is conveniently connected to equipment with a 0.25 V input sensitivity.

The response of various energy β-emitters to amplification, discriminator setting, voltage and detector thickness is presented. The major effects of thermionic emissions and external cooling on background levels are reported.

A method of measuring the concentration of argon-41 in air. L. A. TURNER, W. R. CHAMPION and J. A. MOHRBACHER, Lockheed Aircraft Corp., Dawsonville, Georgia.

Procedures and apparatus for the determination of the concentration of argon-41 in air are described. The method consists of the collection of a large volume of air, removal of essentially all of the constituents, except argon, and counting the residual argon in a high efficiency counter. The apparatus presently in use produces a count rate of twice background with a concentration of approximately 10^{-16} c of argon-41 per cm^3 of original sample.

An estimation of radiation exposure from fallout. R. SZEPKE, Central Laboratory for Radiological Protection, Warsaw, Poland.

The exposure dosage for the population of Middle Europe, due to fallout (Sr^{90}, Cs^{137}, and C^{14}), is discussed and calculated, including geographical distribution, stratosphere residence, radioactive decay, plant uptake, soil leaching, and dilution of the radionuclides with stable elements in nature.

Children born in the period 1955 through 1958 will receive the greatest internal dose from strontium-90, about 1400 mrem per 30 years. Maximum dose rate is calculated and found to occur in 1961 to 1962. Doses are calculated for the period 1954 through 2000.

Radioactive contaminants in the effluent from the Brookhaven National Laboratory air-cooled reactor. C. F. FOELIX, Brookhaven National Laboratory, Upton, New York.

In addition to a large amount of argon-41, several curies per day of particulate and vaporous activities

resulting from fission or activation are discharged in the cooling air of the BNL air-cooled research reactor. The nature and quantity of this activity has been investigated by a variety of techniques. These techniques and the results obtained will be discussed with particular attention given to estimates of the rate of release of iodine activity and the ratio of iodine activity to non-iodine activity.

Health physics in a state health department. C. E. ROESSLER and U. CLARK, Florida State Board of Health, Jacksonville, Florida.

This is not a technical paper but a report to the Society on the role health physics is playing in one state health department.

Activities include the survey of medical, dental and industrial X-ray installations, consultation to prospective users of radiation, review of plans for X-ray and radioisotope facilities, establishment and operation of programs and laboratories for monitoring of radioactive materials in all phases of the environment and participation in the drafting of comprehensive regulations for radiation control. Special problems arise in the handling of data from mass surveys and statewide registration of radiation users, and in perfection of techniques for mass X-ray safety surveys. Research opportunities that present themselves are epidemiological studies of the effect on health status of variations in background radiation exposure, a study of the factors affecting population dose due to diagnostic X-ray exposure, and a study of the environmental factors affecting population dose due to radioactive materials.

Use of multi-channel analyzers in health physics training. H. F. STEWARD and C. D. GEILKER, Rbt. A. Taft Sanitary Engineering Center, Public Health Service, Cincinnati, Ohio.

γ-Scintillation spectrometers of the multi-channel analyzer type have proved to be a particularly useful tool in teaching certain principles of radiation physics. Hundred channel and 20-channel analyzers have been used during the past three years in instruction of more than 1200 participants in short term courses in Radiological Health. In the basic courses students are introduced to the spectrometer and thereafter proceed to the identification of γ-emitters and determination of half life of specific components of radionuclide mixtures. Experiments in the more advanced courses included determination of X-ray spectra as a function of kilovoltage and filtration, measurement of attenuation coefficients, quantitative analysis for specific radioactive contaminants in drinking water, and measurement of effective half life of radionuclides in laboratory animals.

A shadow shield for whole body counting. W. C. ROESCH and H. E. PALMER, Hanford Atomic Products Operation, Richland, Washington.

The concept of a shadow shield and how its use in whole-body counting reduces the background shielding to a minimum is described. The background in this counter is the same as that obtained in an iron room, except at the energies below 0.5 MeV it is 10 per cent higher due to scattered radiation. A scanning counter technique is also described which when used in conjunction with a shadow shield is satisfactory for measuring the normal radioactivity in the body. The shadow shield counter would be light enough that one could be built in a truck. This would permit the counter to be taken to various work areas or other suitable locations.

Symposium on Dosimetry. Chairman: K. Z. MORGAN, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Some general remarks on radiation dosimetry. G. S. HURST and R. H. RITCHIE, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Since the time of its establishment by the careful work of GRAY, the Bragg–Gray principle has served as the main basis for radiation dosimetry measurements. The Bragg–Gray principle, conceived as a means of measuring the total energy absorbed per gram of material occupying the sensitive region of the radiation detector, has been successfully employed to measure the absorbed dose in various kinds of matter for ionizing radiation over a wide range of energy. However, with the present large scale application of different types of ionizing radiation and the resulting more stringent requirements for the protection of man against these radiations, it is desirable to characterize the interaction of radiation with matter in several ways. Such characterizations are also necessary in the increasingly sophisticated field of radiation biology. New advances in radiation physics make it possible to approach the problem of dose determination in many more ways than were available at the time of the original work of GRAY. Hence, it is desirable to state the basis for radiation dosimetry in a more general way which will provide a more systematic basis for achieving the various dose characterizations referred to above.

This paper shows that, whereas the Bragg-Gray principle permits one to deal only with the total amount of energy absorbed in the radiation detector, with a more general formulation one may deal with various quantities derived from operations on the energy loss distribution resulting from the interaction of radiation with the detector. Examples are shown to illustrate the flexibility of this more general statement, and several new approaches to practical problems are suggested.

The calorimetric measurement of local absorbed dose. J. S. LAUGHLIN, Memorial Center, New York City.

This paper will review the development and results of the application of calorimetric methods to the measurement of the intensity and local absorbed dose produced by X-ray beams. The special needs which existed in the high energy radiation region because of the inadequacy of conventional methods of dose measurement will be discussed. The results obtained with calorimeters for intensity measurements from 200 kV to 22 MeV will be given.

The problem of the determination of the local absorbed

dose will then be described. The development of calorimeters of relatively large amounts of material in which a small segment has been internally isolated will be described. The details and operation of calorimeters constructed of polystyrene, conducted polystyrene, and carbon will be presented, with description of the specific problems and corrections necessary in each case. The application of these calorimeters to cobalt γ-rays and the comparison with extrapolation chamber ionization measurements and radiation chemistry measurements are given.

Pulse counters for γ-dosimetry. A. R. JONES, Atomic Energy of Canada Ltd., Chalk River, Ontario.

The paper discusses the application of pulse counters to the measurement of γ-doses and dose rates. As examples some properties of G–M-Counters and silicon p–n junction detectors are described. Electronic techniques for handling, displaying and storing the digital information are presented. The advantages and limitations of pulse counters, as compared with more common detectors providing analogue information, are discussed.

Solid state and luminescence dosimetry. F. H. ATTIX, U.S. Naval Research Laboratory, Washington, D.C.

Several types of solid state devices have emerged in the field of radiation dosimetry in recent years. The principal ones will be described briefly, including silver-activated phosphate glass, thermoluminescence of CaF_2–Mn, luminescence degradation in organics, photoconductivity in CdS crystals, and others. Recent developments at NRL will be dealt with in more detail, particularly those relating to the CaF_2–Mn dosimeter.

Developments in radiation polymerization dosimetry. F. E. HOECKER, University of Kansas, Lawrence, Kansas.

Recent improvements in the dosimetric liquid have extended the dose range of the radiation polymerization dosimeter from 500 rads (at 50 rads/min) up to the previously reported 10^7 rads. The increased sensitivity has greatly facilitated the study of the properties of the liquid in relation to dose rate and energy dependence. Studies with X-rays, Co^{60} γ-rays, and electron beams reveal negligible energy dependence above 200 kV. Dose rate dependence is insignificant above 2500 rads/min. Data will be presented to show the effects of dose rate dependence below 2500 rads/min.

Important characteristics of this dosimetric system in relation to irreversibility and sharpness of endpoint, light and heat sensitivity in relation to storage, and practical applications in dose control and research will be described.

Absorbed dose and local energy density. H. H. ROSSI, Columbia University, New York City.

When matter is irradiated with ionizing radiation, the absorbed dose is defined as the ratio of E/m where E is the energy absorbed in a mass, m. The energy must be determined for a mass that is sufficiently large so that the resultant ratio is representative for the general region in which it is determined. If m is small, the ratio may frequently not be a representative value, particularly when the absorbed dose is low and the radiation is of high LET. In this case, the ratio is denoted by the symbol Z and there can be a wide distribution of values of Z. A detailed knowledge of this distribution is of considerable importance in both radiobiology and radiation damage studies. Methods of measurements of the Z distribution and general considerations pertaining to the distribution will be discussed.

Symposium on "ROVER" Project. Chairman: H. F. SCHULTE, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

Nuclear heat exchanger rockets. R. S. COOPER, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

The purpose of this paper is to present a brief but general view of nuclear heat exchanger powered rockets. We shall first describe the nature of the engines including discussion of the materials, reactor physics, and propellants as they affect performance. There follows an elementary systems analysis and mission study which evaluate the performance of nuclear rockets including comparisons with other propulsion systems. Finally, some of the problems due to the associated nuclear radiation are briefly evaluated.

Radiation safety in the development and use of nuclear energy for rocket propulsion. W. H. LANGHAM, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

Three prototype nuclear propulsion reactors have been tested by the Los Alamos Scientific Laboratory. Although future reactors will create formidable radiation safety problems, they will be small compared to those of even nominal nuclear weapons.

A vigorous safety program is being conducted concurrently with engineering development such that adequate assurance of safety will be possible by the time flyable reactors are available. Intensive studies, involving both measurements and theoretical predictions, carried on since the inception of the reactor program, have enabled delineation of the problem areas and resulted in reasonably sound predictions of the potential radiation hazards of testing and flying a minimal performance system capable of producing 1000 MW of power for an operating cycle of 5 min.

The reactor, although containing more U^{235} than an atomic bomb, could not produce a significant nuclear explosion under the most drastic accident conditions.

Prompt neutron and γ-radiation during operation will not deliver significant exposure to test and launch crews at a distance of 1 mile. Accidental release of accumulated fission products during static testing, launch pad failure, or early mission abort would be confined largely to the controlled area of the launching or testing site. Impact outside the controlled area of an operated reactor, either as a result of late mission failure or re-entry from orbit,

could produce a potential radiation hazard to the public domain as well as an international political incident of considerable magnitude. Means of dealing with this particular problem area need further study.

General biospheric contamination from nuclear rocket operations is an insignificant problem compared to that created by nuclear weapon tests. Radiation problems of manned nuclear rockets are prodigious but amenable to solution.

KIWI-A-PRIME and KIWI-A-THREE radiation measurement results. H. S. JORDAN, Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

Radiation measurements have been made during tests on three rocket propulsion reactors at the Nevada Test Site. Data will be presented on direct γ- and neutron measurements and on measurements of radiation due to passage of the fission product cloud and fallout from the cloud at various distances. These include the dose from inhaled fission products. All types of exposure have been added together to give total dosage figures as a function of position with reference to the Test Cell.

Symposium on Radiation Accidents. Chairman: E. L. SAENGER, University of Cincinnati.

Graphic aids in determining reactor hazards in the event of an accident. J. C. COUCHMAN, Convair, Fort Worth, Texas.

Convair-Fort Worth has devised many graphic and tabular aids that make possible a quick estimate of environmental hazards resulting from a nuclear reactor accident. These include curves of the reactor buildup of biologically hazardous fission products, tables of experimental release per cents, an atmospheric dispersal model, an analytic expression and curves for determining exposure isopleth areas, tables of disposition velocities, and curves for estimating external dose from cloud passage. These and biological exposure criteria used at Convair for hazards studies are discussed.

Influence of continuing clinical observations on dose estimates one year after a radiation accident. M. INGRAM, J. W. HOWLAND, C. L. HANSEN, JR., H. MERMAGEN and C. R. ANGEL, University of Rochester School of Medicine and Dentistry, Rochester, New York.

Delayed manifestations of X-ray injury in the most severely injured casualty in the Lockport incident include effects on the nervous system, vascular system, testes and skin. Only one of the most seriously injured men has shown prominent late effects. Health of the others corresponds to their respective conditions before exposure. Earlier estimates of dose may be modified in some of the cases on the basis of retrospective analysis of the clinical courses. Clinical data are summarized and modified dose estimates discussed.

The radiation incident at the Sandia Corporation Van de Graaff Facility on 8 November, 1960. H. L. RARRICK, G. E. TUCKER, JR., W. D. BURNETT and W. H. KINGSLEY, Sandia Corporation, Albuquerque, New Mexico.

On 8 November, 1960, a staff member was exposed to an electron beam generated within a Van de Graaff accelerator. He received an incident dose of approximately 36,400 ergs/cm^2 to his face, 10,400 ergs/cm^2 to his eyes, and 202,000 ergs/cm^2 to his hands. The accelerator operator checked the intensity of the beam with hand held survey meters. He received an incident dose of about 20,200 ergs/cm^2 to his right hand and 53 rads to his body. This report lists the events leading to the exposure, the measurements and calculations used to determine the dose, and the depth of dose.

Medical aspects of accidental exposure to a Van de Graaff generator. S. P. BLISS, Sandia Corporation, Albuquerque, New Mexico.

Accidental exposure to high dosage of β-ray presents no great emergency medical problem but does produce permanent damage and disability, severity of which is directly related to dosage. In the case presented, no systemic reaction was observed, all signs and symptoms were localized to areas exposed, and all laboratory results were within normal limits. This case demonstrates the value of out-patient care. Progress of signs and symptoms closely parallel previously reported cases of β-ray burns.

Health physics aspects of the SL–1 accident. J. R. HORAN and W. P. GAMMILL, Idaho Operations Office, U.S.A.E.C., Idaho Falls, Idaho.

An explosion in the SL–1 reactor resulted in the deaths of three persons, damage to the reactor, and high-level radiation and contamination in the reactor room. Data from the environmental monitoring program will be presented. The emergency and health physics procedures employed during the first two weeks of recovery operations will be described. Recommendations will be presented for disaster planning as the result of the SL–1 accident experience.

Analytical problems associated with the SL-1 accident at the National Reactor Test Site. E. R. EBERSOLE, Idaho Operations Office, U.S.A.E.C., Idaho Falls, Idaho.

No short-lived fission products indicative of a nuclear excursion were identified in γ-spectra of early samples. Contamination with aged fission products up to 25 r/hr on all samples made analyses for specific isotopes extremely difficult. Evidence of a nuclear excursion was obtained from gold-198, copper-64, chromium-51, and cobalt-58 activation products. NVT values were estimated from these activities. The number of fissions was estimated from strontium-91 and uranium content of particulate material shaken from clothing.

Health Physics Pergamon Press, 1961. Vol. 6, p. 253. Printed in Northern Ireland.

A LIST OF SOME OF THE PAPERS TO BE PUBLISHED IN FUTURE ISSUES

R. K. Hilliard, C. E. Linderbroth and A. J. Scott: Fission product release from overheated uranium—a laboratory study.

R. A. Harvey: Some trends and recent health physics instrumentation developments at Hanford.

Egilda D. Witherell and Russell F. Cowing: Status of state legislation related to ionizing radiation.

D. G. Ott, W. H. Schweitzer, J. A. Sayeg and P. S. Harris: Trichloroethylene-saturated water and tetrachloroethylene chemical dosimeter systems—neutron response.

Sohei Kondo: Responses of silver-activated phosphate glass to α-, β- and γ-rays and neutrons.

J. L. Wolfson and J. C. Dyment: Dosimetric properties of electrets.

Alan R. Smith: A cobalt neutron-flux integrator.

Nathaniel F. Barr, Mary B. Stark, Joan Hands and John S. Laughlin: Dosimetry with small silver-activated glass rods.

R. E. Alexander: Sea water monitoring following radioactive waste disposal operations.

W. J. Bair, D. H. Willard and L. A. Temple: Plutonium inhalation studies—I. The retention and translocation of inhaled $Pu^{239}O_2$ particles in mice.

V. Herrero Climent: An analysis of the Chicago Synchrocyclotron personnel doses over a six year period.

R. W. Perkins: A large well crystal for the direct measurement of trace amounts of radioisotopes in environmental samples.

Stuart C. Black: Low-level polonium and radiolead analysis.

G. W. Dolphin and I. S. Eve: Some calculations on the acceptable limits for the inhalation of radiostrontium in an emergency.

C. L. Comar, R. H. Wasserman and A. R. Twardock: Secretion of calcium and strontium into milk.

E. E. Pochin and C. F. Barnaby: The effect of pharmacological doses of non-radioactive iodine on the course of radioiodine uptake of the thyroid.

A. S. Chhabra and R. K. Hukkoo: Natural and fallout gamma activity in foodstuffs.

HEALTH PHYSICS SOCIETY
PLACEMENT COLUMN

Orders for ads to be sent prepaid to the Placement Committee at the address below. Ads will appear in the next issue following receipt.

1-Time Charges:

Position wanted: $0.25 per word ($4.00 minimum)
Position offered: $1.00 per word ($15.00 minimum)

Replies to employer or to
Mr. Cecil R. Buchanan,
Chairman Placement Committee H.P. Society,
5725 Rossmore Drive,
Bethesda 14, Maryland.

POSITIONS OFFERED

fee paid positions for
HEALTH PHYSICISTS
Write for Application • No Resume Required • Confidential Handling
We are a national placement service with technical experience, dedicated to people in the nuclear field . . . wherever radioactivity is a factor.
ATOMIC PERSONNEL, INC.
Suite 1207-R, 1518 Walnut Street, Philadelphia 2, Pa.
An Employment Agency for the Nuclear Field

HEALTH PHYSICS TECHNICIANS

To work with engineers and scientists in Sandia's nuclear program. Work with accelerators, a pulse reactor, and a 5 megawatt engineering reactor for radiation effects studies. Some field work may be involved.

At least two years of formal education in physics, mathematics, or engineering plus two years or more of reactor or radiation laboratory experience.

Sandia Corporation, located in Albuquerque, N.M., is engaged in research and development of nuclear weapons and other projects for the AEC. Albuquerque is a modern city of about 225,000; has an excellent climate and many cultural and recreational attractions. Winters are mild, summer nights are cool, and there's plenty of year-round sunshine. Liberal employee benefits include generous vacations, retirement and insurance plans and an educational assistance program. Paid relocation allowance.

Send resume to: General Employment Section 533.

SANDIA CORPORATION
ALBUQUERQUE, NEW MEXICO

RADIOLOGICAL EDUCATION

New position in radiological education needs scientist with strong teaching interest. Responsibilities include design and conduct of courses in technology of radiation protection to meet diverse requirements of major nuclear site. Ph.D. degree, knowledge of radiological physics, personnel exposure control desirable. Salary commensurate with background. Send resume to:

J. M. Atwood, Hanford Laboratories,
General Electric Company, Richland, Washington.

All qualified applicants will be considered for employment regardless of race, color, creed, or national origin.